Measurement of the 2200 m/sec neutron-proton capture cross section*

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The neutron-proton capture cross section at the neutron laboratory velocity of 2200 m/sec has been determined from the time decay of the thermalized neutron population following short bursts of fast neutrons in water samples of widely varying sizes. Use of an intense pulsed neutron beam enables elimination of many of the problems encountered in earlier experiments. The present result for this cross section is 332.6 ± 0.7 mb, which is the most precise of any result obtained by this method and is comparable in accuracy and consistent with the most accurate values determined by any method.

NUCLEAR REACTIONS $p(n, \gamma)d$, $E=0.0253$ eV; measured lifetimes of thermal neutrons in water samples of various sizes; deduced σ_{q} .

I. INTRODUCTION

Among the many experimental and theoretical studies which reveal important aspects of nuclear forces, the most significant have been those centered on the interactions between neutrons and protons. As the radiative capture of slow neutrons by protons is governed by the singlet and triplet forces, the measurements of the capture cross section can lead to important information on effective ranges.

In most light nuclei, the thermal neutron radiative capture cross section, with certain exceptions, varies in the range 1-70 mb. Important among these exceptions is the case of the hydrogen nucleus whose capture cross section is well above 300 mb. This relatively large value prompted Fermi to postulate a photomagnetic capture process, which, since it involves only S states, results in a $1/v$ dependence of the cross section.

The theoretical work for the calculation of $n-p$ capture cross section has ranged from simple phenomenological theory to complex treatments of the interaction effect. All theoretical calculations until 1972 yielded values of $5-10\%$ less than the measured results. However, in that year, two independent papers reported that the major portion of this discrepancy could be accounted for by the inclusion of a one-pion-interaction effect.

There are basically three experimental methods aimed at determining the hydrogen capture cross section for thermal neutrons, depending on the type of the neutron source used: steady state, oscillatory, or pulsed.

In the steady state method, the spatial neutron distribution is measured first in a large hydrogenous medium, and then with a homogeneously distribution is measured first in a large hydro-
genous medium, and then with a homogeneously
distributed standard absorber, such as boron.^{1,2} This technique, also known as the cross section

ratio technique, is, however, limited by the accuracy of the value of the boron cross section and the knowledge of the ¹⁰B enrichment. Another variety of the steady state method is the diffusion length experiment which consists of measuring the relaxation length of thermal neutrons along the central longitudinal axis in a moderating medium whose transport cross section, however, must be known. '

In the oscillatory method, the response of a critical pile to a periodically varying absorber is determined. This technique is a relative method and, as in the case of the cross section ratio technique, requires standard absorbers of known cross sections.^{4,5} nique, requires standard absorbers of known cross sections.^{4,5}

The first measurement with pulsed neutron sources employed a large water sample to measure the mean life of thermal neutrons. This technique was initiated by Manley, Haworth, and Luebke,⁶ was initiated by mailey, haw of the and Ede
followed by von Dardel and Waltner,⁷ Meads $et al.,⁸$ and most recently by $Cox,$ Wynchank, and Collie,⁹ who claimed the highest accuracy of all determinations to date. In this method, the decay of the entire neutron population, integrated over the whole volume, is determined. This method has the advantage of eliminating the requirement that higher harmonics of the neutron flux distribution be negligible. However, corrections must still be made for the finite size of the volume over which measurements are made.

In the usual pulsed neutron source method, developed independently by von Dardel and Sjoveloped independently by von Dardel and Sjottrand, ¹⁰ Scott, Thomson, and Wright, ¹¹ and Antonov *et al.*, ¹² the capture cross section i Antonov et $al.$, 12 the capture cross section is determined from the rate of exponential decay of the fundamental mode of the thermalized neutron population following short bursts of fast neutrons into several water assemblies of different dimensions. The pulsed neutron source technique affords an

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absolute and direct determination of the capture cross section. It is also known as the neutron lifetime technique, since the decay of neutrons following the pulse yields their lifetime in the particular medium. The limits on sample sizes are set by, among other factors, the neutron source intensity. In general, for a given source intensity, there corresponds a range in sample sizes beyond which the measurement of the fundamental mode decay constant may not be entirely successful. In the extreme cases of small and of large samples, the time interval over which useful data may be collected is reduced considerably. The need to reject the initial part of the decay curve, in the case of a large medium in which the contribution of higher spatial harmonics to the fundamental (mode) persists over a longer period of time, restricts the time interval of the useful data to the relatively low count rate range. This condition may be remedied by the use of a strong source.

These and subsequent experiments by steady state, pile oscillator, and pulsed methods yielded values of the $n-p$ capture cross section in the range 321 to 337 mb. A review of both theory and measurements using pulsed neutrons has beer
given by Cokinos.¹³ given by Cokinos.

In the present measurement, we have used the intense pulsed neutron beam of the Columbia Nevis Synchrocyclotron to measure the exponential decay of thermal neutrons in water samples of a wide range of sizes. The capture cross section has been determined with high precision from the extrapolated infinite medium decay constant of the size-dependent decay function fitted to our data. The present work differs from earlier experiments in that (a) we have used a much higher intensity pulsed neutron source than previously available, which allowed us excellent counting statistics, and which permitted larger sample sizes to be used; (b) a clean pulse was used with negligible neutron production between bursts; (c) an improved time-analyzing apparatus was employed, and (d) no foreign objects were introduced into the samples. In the following sections, the method applied in these experiments is summarized, the experimental procedure is described, and the results are discussed and compared with earlier measurements as well as with theoretical calculations.

II. METHOD OF MEASUREMENT

The theory underlying this measurement has The theory underlying this measurement has
been given by Nelkin.¹⁴ A brief summary, togethe with specifics for the present'measurement, is given here.

In a typical pulsed neutron source experiment,

short bursts of fast neutrons, separated by time intervals long compared with the neutron lifetimes, are introduced into the sample under investigation. Upon colliding with the nuclei of the moderating medium, those neutrons not escaping from the sample slow down to energies in the thermal region. After an equilibrium distribution, both as to thermalization and as to establishment of the fundamental spatial mode, has been attained, the neutron population decays exponentially with a time constant λ given by

$$
\lambda = \lambda_0 + D_0 B^2 - C B^4 \tag{1}
$$

where λ_0 is the infinite medium decay constant, D_0 and C are coefficients related to the diffusion and thermalization properties of neutrons in the medium, respectively, and B^2 is the buckling, a parameter characteristic of the geometry and dimensions of the sample. For a rectangular parallelepiped sample, whose dimensions are a_x , a_y , and a_z , the buckling is given by

$$
B^2 = B_x^2 + B_y^2 + B_z^2 \t\t(2)
$$

where

$$
B_p^2 = [\pi/(a_p + 2d_p)]^2; \quad p = x, y, z
$$

and d_x , d_y , and d_z are the extrapolation distances in the x , y , and z directions, respectively, i.e., the distance beyond the edge of each surface where the solution of the diffusion approximation to the neutron transport equation vanishes. In the present investigation, the extrapolation distances are taken from the work of Schmidt and Gelbard¹⁵ whose results may be fitted by the expression

$$
d_p = d_0 - d_1 B_p^2; \quad p = x, y, z \tag{3}
$$

with $d_0 = 0.335$ and $d_1 = 0.0232$ for slab geometry.

For large slab samples, the extrapolation distance approaches 0.335 cm. Approximately the same value for d_0 was obtained in calculations by Dorning¹⁶ for the case of large spheres. In addition, d_0 can be calculated from D_0 , and, after averaging over the thermal neutron distribution, gives approximately the same value as above. Thus the value of d_0 used here is well founded and independent of geometry. It is considered to have an uncertainty of 2% .

The buckling-dependent term, d_1B^2 , becomes more important for the smaller size samples. Dorning's¹⁶ calculations for spheres are presented in a different manner than in the form of Eq. (3), but when recalculated give a value for d_1 not too different from that used here. It is estimated that d_1 is known to better than 30%.

It will be seen below that these uncertainties in d_0 and d_1 have a negligible effect on the results of the present measurements.

For a $1/v$ absorber, as in the case for water,

$$
\lambda_0 = Nv \sigma_a \,, \tag{4}
$$

where v is the neutron velocity, N is the number of molecules per cm³ in the sample, and σ_a is the absorption cross section of the molecule. Thus, the determination of λ_0 leads to the value of σ_a at the standard neutron velocity of 2200 m/sec when v is set equal to that value. Hence, the experimental aspect of the measurement consists of determining the decay constant λ for samples of water of different sizes (i.e., different values of the buckling). λ_0 is then determined by a best fit of the data to Eq. (1), and the capture cross section of hydrogen is calculated from Eq. (4) after a, small correction for the capture cross section of oxygen.

III. EXPERIMENTAL ARRANGEMENTS

The experiments presented in this paper have been carried out with Columbia University's Nevis Synchrocyclotron used as an intense pulsed neutron source. Although the cyclotron is primarily used as a high energy physics tool with its ability to produce protons in the hundreds of MeV range, it is also a powerful accelerator-based neutron source. A description of the experimental setup is given in the following sections.

A. Neutron source

The Nevis Synchrocyclotron built in the early 1950's to produce 385 MeV protons for high energy physics research has been developed by Havens and Rainwater^{17,18} into a high intensity pulsed neutron source for a velocity spectrometer. In this machine, the proton beam is deflected by two graphite electrodes and is allowed to strike a lead target. Each 385 MeV proton produces about 10 evaporation neutrons. These neutrons have a Maxwellian distribution of energies mainly under 10 MeV. At a beam current of \sim 0.4 μ A, the instantaneous neutron intensity is 2×10^{19} neutrons/ sec. Some of the evaporation neutrons are moderated in a water box below the lead target and a spectrum of partially moderated neutrons is produced with some neutrons emitted in the direction of the flight path. During these experiments, the modulation frequency of the cyclotron was 70 Hz giving 70 bursts of neutrons per second, each of about 25 nsec duration.

B. Sample assembly

Figure 1 shows a cross section of the box used to isolate the sample from the surrounding media. The 2 cm thick boron carbide slab facing the neu-

FIG. 1. Cross section view of the box housing the samples and the detectors.

tron beam becomes opaque to neutrons below about 50 eV, thus limiting the time during which neutrons enter the sample. The interior of the chamber was kept free of any hydrogenous material which might influence the decay constant. The sample assembly was located 6 m from the neutron source.

C. Samples

The geometry of the water samples used consisted of parallelepipeds of square cross sections ranging in dimensions from 4.45 to 35.5 cm and of various heights. A broad range of bucklings was thus obtained. The samples under investigation were held in containers made of 2S aluminum 0.16 cm thick.

Distilled water has been used in all measurements. Care was taken to ensure that no air bubbles were introduced into samples under study. It is noted here that, unlike some previous experiments, all samples were kept free of any foreign objects, such as detectors and target hardware, that normally tend to distort the neutron flux distribution.

D. Detector and electronics

Thermal neutrons were detected with 2.54 cm diameter, 25 cm long ³He filled proportional counters. The detectors were housed in a specially shielded emplacement directly below the sample, such that the detector was part of the "floor" on which the sample rested. In the case of small samples, the portion of the detector extending beyond the area of the sample was covered with cadmium so that only neutrons diffusing from the bottom of the sample were counted. In the case of

the larger samples, higher harmonic contamination was reduced by the use of cadmium sheets with sections cut out to expose the detector to only those portions of the sample corresponding to the first zero of the harmonic above the fundamental.

The amplified and shaped detector pulses were \det into a time-of-flight analyzer which was speci-
ally designed and built for these experiments.¹⁹ ally designed and built for these experiments.¹⁹ The analyzer's basic features are channel widths of 0.5 μ sec, 1 μ sec, or 2 μ sec, and a data collecting interval extending to 7 msec. The time-offlight system was interfaced to a PDP-8 on-line computer for the storage of data, for display, and for calculational and output operations. This system allowed flexibility in the acquisition and handling of data. The only dead time limitation was that no more than one count could be stored in one time channel per neutron burst, an insignificant effect on the data used in the analysis. In order to verify the day-to-day stability of the electronics as well as to check reproducibility, measurements on selected samples were made at regular intervals.

IV. ANALYSIS, RESULTS AND DISCUSSION

A. n-p capture cross section

The immediate results of the measurements were, for each sample of water, sets of count rates as a function of the time from the neutron burst. The analysis of these data proceeded in two steps: (1) For each sample configuration, the asymptotic decay constant of the neutron population was determined by selecting first that portion of the count rate vs time plot for which the fundamental mode had been established and then determining the decay constant by a weighted least squares fit to an exponential function, and (2) B^2 was calculated for each sample by use of Eqs. (2) and (3) and a weighted least squares fit made to Eq. (1). σ_a was then calculated using Eq. (4). (The weighting used in the least squares fits was inversely proportional to the square of the statistical error at each point.)

In the initial fit to Eq. (1) , 3 out of a total of 17 points were found to deviate excessively (4 standard deviations) from the best fit parabola so as to give a poor fit on the basis of the χ^2 criterion. The least squares fit to Eq. (1) was redone without these three data points and resulted in a value of λ_0 of $4882 \pm 10 \text{ sec}^{-1}$, which corresponds to a thermal neutron lifetime of 204.8 ± 0.4 µsec. The χ^2 criterion indicated that the data fit the parabola of Eq. (1) at the 95% confidence level. The value of λ_0 obtained by using all 17 points deviated from the given value by about 1 standard deviation. The data points are shown in Fig. 2, together with the best fit curve in the form $(\lambda - \lambda_0)/B^2$ vs B^2 , which

FIG. 2. Experimental results plotted as $(\lambda - \lambda_0)/B^2$ $v s B²$ where λ_0 is taken as 4882 sec⁻¹. " \times " indicates those measurements deviating by 4 or more standard deviations from the best fit line which has been calculated without these points. " \blacksquare " indicates results for those samples which are near cubical shape and used in an alternate calculation.

allows the statistical errors of the individual decay constants to be clearly evident.

Additional least squares fits to Eq. (1) were performed to check the stability of the above result to variations in the calculational procedure: (a) The coefficients in the expression for the extrapolation distances given by Eq. (3) were varied by the uncertainties stated above (the constant term by 2% and the buckling-dependent term by $30\%)$.

(b) Those samples of near cubical shape were selected for the calculation, to minimize further the influence of the extrapolation distance. (c) A B^6 term was added to Eq. (1). In all of these variations, the change in λ_0 was substantially less than the statistical uncertainty in λ_0 so that no significant error is introduced by the initial procedure used.

Equation (4) together with the quantities given in Table I yielded, finally, the value of the neutron-proton capture cross section at 2200 m/sec as 332.6 ± 0.7 mb.

The following possible sources of error have been examined and are considered to make a negligible contribution to the statistical error cited; (1) impurities in the sample, including dissolved nitrogen from atmospheric air, (2) the effect of the aluminum containers, and (3) errors in the constants of Table II.

The assigned error is thus of a statistical nature only, and there are no known errors which would increase this value.

The value of D_0 and C resulting from the present measurements are to be subjects of a separat
paper.²² However, as another indication of th paper. However, as another indication of the

Constant	Value	Reference This work	
Infinite medium decay constant	$4882 \pm 10 \text{ sec}^{-1}$		
Thermal neutron lifetime	$204.7 \pm 0.4 \text{ m/sec}$	This work	
Thermal neutron speed	2200 m/sec	Standard	
Avogadro's number	$(0.602257 \pm 0.000009) \times 10^{24}$	20	
Water density at 22°C	0.9978 g/cm^3	20	
Molecular weight of H_2O	18.01534 g/g mole	20	
Oxygen capture cross section	0.178 ± 0.025 mb	21	
Temperature	22° C		

TABLE I. Constants needed to evaluate σ_{nb} .

reliability of these measurements, the values of D_0 and C obtained here are in excellent agreement with recent rigorous calculations¹⁶ of the same quantities for water.

B. Comparison with earlier results

Table II summarizes measurements of the neutron-proton capture cross section at 2200 m/sec. The methods used were discussed above and are indicated in Table II.

These results may be grouped into three broad categories: (1) the two with the highest precision, ≤ 1 mb: the present one and that of Cox et al.⁹; (2) a number of results of considerably less accuracy, 5 mb or less, but having values in the range 321 to 329 mb and deviating from the present results by more than 2 standard deviations; and (3) values with much larger uncertainties, together with those of moderate accuracy, which overlap the present results by less than 2 standard deviations. This last group will not be discussed further.

the rast group will not be discussed further.
Cox *et al*.⁹ have obtained a value of 334.2 ± 0.5 mb, the best accuracy claimed up until the present time, using a different pulsed source technique, as described above.

Since two forms of the pulsed neutron measurements have yielded the results of highest accuracy, it is appropriate to contrast the two methods. In both methods the capture cross section is in-

TABLE II. Absorption cross section of hydrogen at 2200 m/sec.

Method	σ_a^H (mb)	Year	÷ Author
Pulsed, fixed large sample	330 ± 20	1942	Manley et al. (Ref. 6)
Stationary, σ_R/σ_H	334 ± 5	1947	Whitehouse and Graham (Ref. 11)
Stationary, σ_{R}/σ_{H}	329 ± 4	1953	Hammermesch et al. (Ref. 2)
Pile oscillator	334 ± 7	1953	Harris et al. (Ref. 23)
Pulsed, fixed large sample	$321 \pm 5^{\ a}$	1953	von Dardel and Waltner (Ref. 7)
Pulsed, variable size samples	$323 \pm 8^{\mathrm{b}}$	1954	Scott <i>et al.</i> (Ref. 11)
Pulsed, variable size samples	333 ± 3	1954	von Dardel and Sjostrand (Ref. 10)
Pulsed, variable size samples	329 ± 9	1955	Antonov et al. (Ref. 12)
Pulsed, variable size samples	337 ± 10	1956	Bracci and Coceva (Ref. 24)
Pulsed, fixed large sample	335 ± 4	1956	Meads et al. (Ref. 8)
Pile oscillator	329 ± 2	1957	Cummins (Ref. 25)
Diffusion length	330 ± 4	1958	Dio $(Ref. 3)$
Stationary, σ_{R}/σ_{H}	328 ± 4	1958	Baker (Ref. 26)
Pulsed, variable size samples	337 ± 5 °	1959	Sjostrand <i>et al.</i> (Ref. 27)
Random pulses, large fixed sample	330 ± 8	1959	Stooksberry and Crouch (Ref. 28)
Pulsed, variable size samples	335 ± 5	1961	Meadows and Whalen (Ref. 29).
Pulsed, variable size samples	325.3 ± 1.6	1962	Lopez and Beyster (Ref. 30)
Pulsed, large fixed sample	334.2 ± 0.5	1965	$\cos et al.$ (Ref. 9)
Pulsed, variable size samples	333.1 ± 3.0	1965	Pal <i>et al</i> . (Ref. 31)
Pulsed, variable size samples	335.7 ± 2.7	1965	Arai and Kuchle (Ref. 32)
Pulsed, variable size samples	321 ± 2.5	1969	Nassar and Murphy (Ref. 33)
Pulsed, variable size samples	$323 + 4.7$	1969	Nassar and Murphy (Ref. 33)
Pulsed, variable size samples	332.6 ± 0.7		This work

aNot corrected for flux distortion caused by detectors.

b Not corrected for higher harmonic effects.

^c Hydrogen cross section obtained from polyethylene, CH₂, using $\sigma_c = 3.4$ mb.

ferred from the estimation of neutron lifetime in an infinite medium, since in such a medium, neutrons can disappear only by capture. In the present experiment, the infinite medium case is approached by a series of successively larger samples (smaller bucklings) with corresponding reduction in the leakage of neutrons through the surface. Extrapolation to zero buckling then gives results for the infinite medium. The Cox et al. experiment starts with a localized burst of neutrons in a very large medium with no leakage as a first approximation and observes the decay of the total neutron population.

The main intrinsic difficulty of the method which uses several sample sizes is that a fundamental decay mode must be reached before lifetime measurements can be made. For successively larger samples, this waiting time gets longer and limits the maximum size sample that can be used. In the present experiment, the problem is made negligible by the high intensity of the pulse so that after the required waiting time there is still an adequate neutron population left in the medium for an accurate determination of lifetime. An additional problem arises from possible uncertainties in the extrapolation distances. As discussed above, this is not significant here, but may become important if still greater accuracy is sought.

The main intrinsic problem of the Cox et al. measurements is that the medium used, though large, cannot be infinite, and measurements cannot be made right out to the edge of the medium. Thus, part of the observed decay of the neutron population is due to the leakage of neutrons out of the region where measurements are made, and corrections have to be made for this leakage. There appears to be no estimate of the error introduced in making this correction. Additional experimental problems also arise from the presence of a beam tube and a detector placed inside the medium. The effects of these were investigated to some extent, but not to the, same precision as the claimed final accuracy.

The difference between the present result and that of $\cos \theta t$ al.⁹ is 1.6 mb, which is just twice the square root of the sum of the squares of the individual errors, so that the two results are essentially in agreement.

Turning now to the "low" results of moderate accuracy, it may be seen that there are wide discrepancies among various values of the cross section even among results obtained by the same technique. The general disagreement may be laid to a variety of factors, such as (a) analysis of the data in different ways; (b) failure to establish a fundamental mode of decay; (c) time-dependent background arising from the backscattering of

neutrons from surrounding media; (d) oversimplifications in the treatment of anomalous experimental conditions.

In many of the pulsed source experiments, the detector had been placed inside the sample under investigation. This procedure can cause a distortion in the neutron distribution, depending upon the size, type, and position of the detector in the sample. Possible systematic errors may also arise in measurements performed with the beam tube and target inside the sample. This situation is encountered in experiments with large samples.

We now consider some of the individual measurements yielding these "low" results.

Von Dardel and Waltner' obtained a value of 321 $± 5 mb.$ However, in a subsequent paper, von Dardel and Sjostrand¹⁰ attributed this low value to a time-dependent background whereas a constant background had been assumed in the analysis. retain the analysis.
Lopez and Beyster,³⁰ using a method similar to

the present one, extended their measurements to larger samples as well, and obtained a 325.2 ± 1.6 mb. Since the larger samples gave rise to unattenuated higher harmonics, these authors corrected their decay constants for these higher harmonics. Because of the approximate nature of the correction scheme, this procedure may have resulted in an overcorrection, thus producing too low a cross section value.

Nassar and Murphy³³ reported a value of 321 \pm 2.5 mb based on the same method. Upon fitting their data over a slightly smaller buckling range, these authors obtained an increase in their cross section to 323 ± 4.7 mb. However, their measurements were conducted with a BF₃ detector sealed in a thin Lucite tube and placed inside the samples, which may haVe affected their results.

C. Comparison with theory

Because of the important role of the capture cross section on effective ranges, many attempts were made to compute the thermal neutron-proton capture cross section from basic principles running parallel with the experimental determinations. Until 1972, all theoretical estimates of the thermal neutron capture by protons were about 5 to. 10% lower than the measured values. The difference between the experimental and theoretical values was generally attributed to an "interaction effect," a measure of the distortion of the magnetic properties of the free neutron and proton caused by the nuclear interaction.

used by the nuclear interaction.
The best of these calculations,^{34,35} using the usual single-particle magnetic moment operators with the best known values of effective ranges, gave 302.5 ± 4.0 mb, $\sim 9.5\%$ below the results of

 $\cos et al.$ and those of the present paper.

In 1970, Adler, Chertok, and Miller³⁶ apparently took a step in the right direction by calculating the effects of mesonic exchange currents, using a phenomenological method and nonrelativistic wave functions. However, they obtained an increase in cross section of only $2-3\%$.

Figure 1.13 and September 2.13 and subsequent

In 1972, Riska and Brown,³⁷ and subsequent

ari and Huffman,³⁸ reported that an exchange Gari and Huffman,³⁸ reported that an exchange current correction of about 10% can arise in a straightforward way from one-pion-exchange terms. These corrections can vary by about 1% depending upon the particular potential used in the depending upon the particular potential used in the calculations. In 1976, Simon *et al*.³⁹ found that the inclusion of pion-exchange currents was necessary to account for the electrodisintegration of the deuteron in the low and intermediate momentum transfer region.

Thus the major portion of the discrepancies between experimental and theoretical results appears to have been resolved. The 10% correction supports values near the present one as opposed to the lower values near 325 mb. However, the theoretical calculations are still not sufficiently

accurate so as to predict the 2200 m/sec neutronproton capture cross section with the same accuracy as the experimental results. Theory still needs to incorporate some of the small second order terms which have been neglected.

D. Conclusions

 332.6 ± 0.7 mb has been obtained as the 2200 m/ sec value for the neutron-proton capture cross section. This value has a much smaller uncertainty than all other determinations except for the ± 0.5 mb claimed by Cox *et al.* Since these two "best" values are in essential agreement, and since they were based on two different techniques with different possible sources of error, their agreement implies that the "low" values obtained by others are probably in error, or have uncertainties much larger than claimed.

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