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Neutron mass: Measurement of the $^1H(n,\gamma)^2H$ γ ray and revised values for selected neutron binding energies

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The $^1H(n,\gamma)^2H$ reaction γ-ray energy was obtained from measurement of the energy difference between it and the 2185-keV γ ray emitted in the β^- decay of ^{144}Ce-^{144}Pr. Based on the γ-ray energy value of 2223.247 ± 0.017 keV obtained from this measurement, a value of 2224.564 ± 0.017 keV was determined for the binding energy of the deuteron $S_n(^2H)$. By combining this binding energy value with published mass differences of doublets involving neighboring isotopes and ^1H and ^2H, values of 6257.268 ± 0.024, 4946.329 ± 0.024, 8176.483 ± 0.040, and 10833.297 ± 0.038 keV were obtained for the neutron binding energies of ^3H, ^{13}C, ^{14}C, and ^{15}N, respectively, together with a value of 8071.367 ± 0.029 keV for the neutron mass excess $n - 1$.

[NUCLEAR REACTIONS $^1H(n,\gamma)$, E = thermal; measured E_γ. Deduced neutron binding energies for ^2H, ^3H, ^{13}C, ^{14}C, and ^{15}N; deduce neutron mass. Natural targets, Ge semiconductor detectors.]

I. INTRODUCTION

Precise knowledge of the deuteron binding energy, $S_n(^2H)$ or $S_p(^2H)$, is of importance in a number of areas in nuclear physics. At the most basic level it is one of the measurable parameters which characterize the two-body nuclear interaction. Furthermore, because of the improved accuracy achieved in recent years for mass difference measurements,[1] especially using the radio frequency mass spectrometer developed by Smith,[2-4] the combination of the $S_n(^2H)$ value with mass difference values of selected doublets provides the most accurate value for the mass of the neutron and for the neutron binding energies of a number of light-mass nuclides.[5,6] Neutron binding energies are obtained in this manner by employing mass differences of doublets involving neighboring isotopes together with ^1H and ^2H using the following relationships:

$$^AZ^2H - {}^{A+1}Z^1H = S_n(^{A+1}Z) - S_n(^2H) \quad (1)$$

or

$$^AZ^1H - {}^{A+1}Z = S_n(^{A+1}Z) - [n - {}^1H] , \quad (2)$$

where

$$n - {}^1H = S_n(^2H) - [^1H_2 - {}^2H] . \quad (3)$$

In these relationships the atomic mass denoted by ^AZ is for the neutral atomic of the isotope with atomic number Z and mass number A in its nuclear and atomic ground states.

In the most favorable cases such doublet mass differences have been measured with total uncertainties of less than 20 eV.[3,7] The value of the deuteron binding energy is, at the present time, most accurately obtained from measurement of the energy of the prompt γ ray emitted by the $^1H(n,\gamma)^2H$ reaction with slow neutrons. Currently, the uncertainty of 30 eV quoted on the adjusted value of $S_n(^2H)$[1] is the dominant contribution to the total uncertainties of several of the neutron binding energy values determined using doublet mass differences. Furthermore, even this uncertainty estimate for $S_n(^2H)$ may be unrealistic since the energy values with the smallest error[8-10] included in the adjustment[1] involved analyses of γ-ray spectra measured with Ge(Li) detectors containing mixed full-energy (FE) and double-escape (DE) peaks, and in one case[8] single-escape (SE) peaks. Since there were no corrections made in any of these experiments for the shifts induced between the different types of peaks by the electric fields in the detectors[11] (three planar[9,10] and one closed-ended coaxial[8]), or for the possibility that the FE-DE peak separations might not truly be $2m_0c^2$ as discussed in Ref. 12, additional error components might reasonably be expected to occur. As an example, extrapolating from the electric field effect data shown in Ref. 11 to the planar detectors used in Ref. 9 with field gradients of ~100 V/mm, we would estimate that the additional field effect correction to the $S_n(^2H)$ value quoted in Ref. 9 should be ~40 eV. Since the orientation

of these earlier detectors is now uncertain, the sign of the correction term is indeterminate.

In view of the considerations noted above, coupled with the recent availability of precise sets of γ-ray energies up to 3.5 MeV from several radionuclides,[12] based in a traceable manner to the ^{198}Au γ-ray energy value of 411.804 41 keV reported by Kessler *et al.*,[13] a remeasurement of the prompt γ-ray energy from the $^1H(n,\gamma)$ reaction appeared to us to be appropriate. Preliminary results from this present work have been reported in Refs. 14 and 15.

Concurrent with the present work, a redetermination of the deuteron binding energy, also based upon the measurement of the prompt γ-ray energy from the $^1H(n,\gamma)$ reaction, was reported by Vylov *et al.*[16] Their result is quite discrepant with the present result, and the origin of this discrepancy will be discussed in Sec. III.

II. MEASUREMENT OF $S_n(^2H)$

A small Lucite target placed in an external beam of well-thermalized neutrons from the High Flux Beam Reactor at Brookhaven National Laboratory served as the source of $^1H(n,\gamma)$ reaction γ rays. The thermal neutrons were obtained from the H1B beam channel through two Bi single crystals,[17] each 14 cm long with their 111 orientation parallel to the beam. The transmitted beam was collimated down to ~1.2 cm diameter at the sample position using a borated polyethylene collimator. Gamma rays were detected in either a 23-cm^3 intrinsic planar Ge or a 55-cm^3 true coaxial Ge(Li) detector coupled to a TC 205 linear amplifier, a NS 623 analog-to-digital converter, and a SDS 910 computer for storage of 4096-channel pulse-height spectra. The Ge semiconductor detector was shielded from scattered neutrons and background γ rays by a Bi shield surrounding the detector, and a 7.6 cm diameter $\times$3.0 g/cm^2 thick ^{6}LiH filter interposed between the sample and the detector.

Energy calibration for the $^1H(n,\gamma)$ spectra were obtained by simultaneously recording in these spectra γ rays emitted by radionuclide sources of ^{60}Co, ^{144}Ce, and ^{228}Th. In order not to increase the background, these radionuclide sources were located just outside the neutron beam. Compensation for the effect of source geometry on peak position[11] was achieved by alternating runs with the ^{144}Ce source either closer to or further away from the detector than the Lucite target.

Following the experimental procedures discussed extensively in Refs. 12, 18, and 19 for system nonlinearity corrections, peak position analysis using the GAUSS V computer program,[20] and γ-ray energy calibration, we have measured the energy difference between the 2223-$^1H(n,\gamma)$ and the 2185-^{144}Ce γ rays. These results are summarized in Table I where column 1 gives the detector used, column 2 gives the number of measurements made with each detector, and column 3 gives the weighed average of the difference values measured with each detector system. The total average energy difference, obtained by averaging all of the original measured values, is given in column 5, together with the reduced-χ^2 value, ϵ^2, associated with this averaging procedure.

Using the total average energy difference given in Table I, together with a value of 2185.662 keV [with $\sigma_m = 4$ eV and $\sigma_t = 7$ eV, where σ_m and σ_t are the measurement and total uncertainties (1σ) as defined in Ref. 21] for the energy of the ^{144}Ce γ ray,[12] we obtain the following value for the energy of the $^1H(n,\gamma)$ reaction γ ray:

$$E_\gamma(^2H) = 2223.247 \text{ keV} \quad (\sigma_m = 16 \text{ eV}, \ \sigma_t = 17 \text{ eV}).$$

In order to convert this γ-ray energy into a transition energy, which for slow neutron capture is equivalent to $S_n(^2H)$, it is necessary to correct for the energy of recoil of the deuteron produced in the reaction. This recoil energy is computed to be 1.317 keV based on the 1973 adjusted values of the fundamental constants.[22] Thus, applying this correction, we obtain the following value for the binding energy of the deuteron:

$$S_n(^2H) = 2224.564 \text{ keV} \quad (\sigma_m = 16 \text{ eV}, \ \sigma_t = 17 \text{ eV}).$$

This value is significantly lower than the adjusted value for $S_n(^2H)$ given in Ref. 1. Furthermore, examination of the source data[8-10] for the adjusted value indicates that it is primarily based on the ^{60}Co γ-ray energies as calibration. Comparison of

TABLE I. Measured 2223 [$^1H(n,\gamma)$] – 2185 [^{144}Ce] energy difference.

Detector	Number of measurements	Energy difference [a] (eV)	Average energy difference ϵ^2	Value [a] (eV)
BNL-2	5	37 603(24)	0.2	37 585(15)
OR-15	11	37 575(19)		

[a] The uncertainty (in the least significant digits) is shown in parentheses following the difference energy.

TABLE II. Comparison of the calibration energies used by Vylov *et al.* (Ref. 16) to determine the energy of the $^1H(n,\gamma)$ γ ray [corrected to a value of 411.804 41 keV for the energy of the 411-^{198}Au γ ray (Ref. 13)] with those reported in Ref. 12.

Calibration set[a]	Isotope	γ-ray energy (keV) Vylov *et al.*[a]	γ-ray energy (keV) Ref. 12	Difference (Vylov—Ref. 12) (ppm)
Fig. 3	^{88}Y	1836.120	1836.063	31
	^{144}Ce	2185.662	2185.662	0
	^{24}Na	2754.059	2754.030	10
Fig. 4	^{56}Co	1771.392	1771.350	24
	^{88}Y	1836.120	1836.063	31
	^{56}Co	2015.247	2015.179	34
		2034.823	2034.759	32
		2598.510	2598.460	19

[a]Reference 16.

the values used for the energies of the ^{60}Co γ rays in these earlier works[8-10] with the present values[12,21] would suggest that the adjusted $S_n(^2H)$ of Ref. 1 should be increased by ~10 ppm to a value of 2224.650 keV. This increases the discrepancy to 86 eV (39 ppm). As we have discussed in the previous section, it is not unreasonable to attribute a discrepancy of this magnitude to the electric field shifts between FE and DE peaks present in all of the earlier measurements with Ge semiconductor detectors.[8-10]

Concurrent with the present work, Vylov *et al.*[16] have also remeasured the energy for the $^1H(n,\gamma)$ γ ray and based on this they report a value of 2224.572 ± 0.040 keV for $S_n(^2H)$, which when corrected to a value of 411.804 41 keV for the 411-^{198}Au reference energy standard[13] yields a revised binding energy of 2224.628 ± 0.016 keV. This is 64 eV (29 ppm) higher than the value determined in the present work. In their work, Vylov *et al.*[16] employed two separate energy calibration schemes; one using ^{88}Y, ^{144}Ce, and ^{24}Na (their Fig. 3) and the other using ^{56}Co (their Fig. 4). A comparison of the calibration energy values used by Vylov *et al.*,[16] corrected to current value for the 411-^{198}Au reference energy standard,[13] with those used in the present work is shown in Table II. The calibration energy sets of Vylov *et al.*[16] and the present work are clearly discrepant at an energy of 2223 keV; in the Fig. 3 set by perhaps as much as 10 ppm, and in the Fig. 4 set by ~28 ppm. The weighting employed by Vylov *et al.*[16] for their two sets is unclear, but assuming a straight averaging we can account for ~19 ppm out of the total 29 ppm discrepancy between the $S_n(^2H)$ values of Vylov *et al.*[16] and the present work, solely on the basis of the different values used for the calibration energies. A final comment in regard to the measurements of Vylov *et al.*[16] is that it is not clear from their paper whether they have accounted for shifts in peak positions that might have occured as a result of the electric field effect,[11] due to the extended nature of their $^1H(n,\gamma)$ source as compared to the presumably point radionuclide calibration sources.

III. THE NEUTRON MASS

The mass of the neutron is currently most accurately determined using Eq. (3) to compute the neutron-hydrogen atom mass difference. In that equation both $S_n(^2H)$, which we have measured in the present work, and the mass difference $^1H_2 - ^2H$ are required.

There have been numerous measurements of the $^1H_2 - ^2H$ mass difference, as summarized in Ref. 1, but by far the most accurate, and the only data of comparable accuracy to the present $S_n(^2H)$ value are those reported by Smith[3] and Smith and Wapstra.[7] These latter data have been included in the 1977 Atomic Mass Evaluation of Wapstra and Bos,[1] but for the reasons cited in that evaluation their absolute uncertainties were increased by a factor of 2.5. There were no neutron binding energies, except for $S_n(^2H)$, included in that adjustment which had uncertainties comparable to the mass difference values quoted in Refs. 3 and 6. Thus the Wapstra and Bos[1] adjusted value of the $^1H_2 - ^2H$ mass difference depends solely on the adjustment of the total body of mass difference data, and does not include any γ-ray data through Eqs. (2) and (3). We have therefore chosen to use their adjusted mass difference value of 1548.287 ± 0.006 μu to compute a value for $n - ^1H$ from Eq. (3). Applying a mass-energy conversion factor of 1 u = 931 501.6 ± 2.6 keV from the 1973 adjustment of the Fundamental Constants,[22] this converts to an energy of 1442.232 ± 0.007 keV for the adjusted

TABLE III. Variance-covariance and correlation coefficient matrix used to compute the reference uncertainties σ_r of energies (in eV) for mixed 411-^{198}Au wavelength-based and mass-based scales. Here, C_λ and C_M are the conversion factors from m^{-1} to eV and u to eV, respectively. Following Table 33.4 of Ref. 22, the variances and covariances are in $(ppm)^2$ and the correlation coefficient is given below the diagonal.

	C_λ	C_M
C_λ	6.835	6.014
C_M	0.811	8.045

$^1H_2 - {}^2H$ mass difference. Using this mass difference together with our present value for $S_n(^2H)$ in Eq. (3), we obtain the following value for the neutron-hydrogen atom mass difference:

$$n - {}^1H = 782.332 \text{ keV}$$

$$(\sigma_m = 17 \text{ eV},\ \sigma_r = 3.4 \text{ eV},\ \sigma_t = 17 \text{ eV}),$$

where σ_r refers to the reference scale error.

The neutron mass excess can most simply be obtained by rearranging Eq. (3) as follows:

$$n - 1 = S_n(^2H) - [{}^1H_2 - {}^2H] + [{}^1H - 1]. \quad (4)$$

The adjusted value of the ^1H mass excess depends solely on the adjustment of the mass difference data, and not on γ-ray data, as discussed above. We have therefore chosen to use this adjusted $^1H - 1$ value of 7825.037 ± 0.010 μu, or 7289.034 ± 0.023 keV, in Eq. (4) to obtain the following value for the neutron mass excess:

$$n - 1 = 8071.367 \text{ keV}$$

$$(\sigma_m = 19 \text{ eV},\ \sigma_r = 22 \text{ eV},\ \sigma_t = 29 \text{ eV})$$

or

$$n - 1 = 8664.898\ \mu\text{u}$$

$$(\sigma_m = 21 \text{ nu},\ \sigma_r = 4.0 \text{ nu},\ \sigma_t = 21 \text{ nu}).$$

In computing the uncertainties associated with the values of $n - {}^1H$ and $n - 1$ above, and with the values of $S_n({}^ZA)$ discussed in the following section, it must be recognized that the final energies (or masses) are linear combinations of both 411-^{198}Au wavelength-based and mass-based numbers. In order to compute correctly the reference uncertainties associated with such mixed-scale values, it is necessary to utilize the variance-covariance matrix terms associated with the 1973 adjustment of the Fundamental Constants,[22] recognizing that the relevant scale conversion factors can be expressed as follows:

$$1 \text{ m}^{-1} = \left\{\frac{he}{c}\right\} \text{ eV}, \quad \text{wavelength conversion}$$

$$1 \text{ u} = \left\{\frac{c^2}{10^3 F}\right\} \text{ eV}, \quad \text{mass conversion}$$

where following Ref. 22, the braces indicate numerical values only. From these scale conversion relationships, and using the procedure outlined by Cohen and Taylor,[22] we have computed a variance-covariance matrix which is applicable for computing realistic estimates of the reference uncertainties in mixed wavelength-energy and mass-energy relationships. This matrix is shown in Table III, where C_λ is the conversion factor from m^{-1} to eV and C_M is the conversion factor from u to eV. The value of 0.811 for the correlation coefficient between these two conversion factors indicates a high degree of correlation. Since the conversion factors are correlated, the reference uncertainty associated with an energy value, E, determined from a linear combination of wavelength (E_λ) and mass (E_M)-based energies is obtained as follows:

$$E = E_\lambda + E_M = (f_\lambda + f_M)E,$$

with

$$f_\lambda = \frac{E_\lambda}{E} \text{ and } f_M = \frac{E_M}{E},$$

we have

$$\sigma_r^2 = \frac{\partial E}{\partial C_\lambda}\frac{\partial E}{\partial C_\lambda}v_{11} + 2\frac{\partial E}{\partial C_\lambda}\frac{\partial E}{\partial C_M}v_{12} + \frac{\partial E}{\partial C_M}\frac{\partial E}{\partial C_M}v_{22},$$

where the v_{ij} are the covariances. If σ_r is in eV, E_λ and E_M in MeV, and the v_{ij} in $(ppm)^2$, this expression reduces to

$$\sigma_r^2 = E_\lambda^2 v_{11} + 2E_\lambda E_M v_{12} + E_M^2 v_{22}$$

or, if σ_r is in ppm, the v_{ij} in $(ppm)^2$, to

$$\sigma_r^2 = f_\lambda^2 v_{11} + 2f_\lambda f_M v_{12} + f_M^2 v_{22}.$$

IV. OTHER NEUTRON BINDING ENERGIES

For several of the light-mass stable nuclei, for which mass differences of doublets involving neighboring isotopes together with ^1H and ^2H have been precisely measured,[7] the relationships expressed by Eqs. (1) and (2) provide by far the most accurate values for the neutron binding energies of the higher mass isotopes $S_n({}^{A+1}Z)$. We have obtained the neutron binding energies listed in Table IV by combining the present value for $S_n(^2H)$ with such doublet mass difference values. In computing these values for $S_n({}^{A+1}Z)$ we have chosen whenever possible, for the reasons discussed earlier in Sec. III, to use the adjusted mass differences and their associated uncertainties from Ref. 1. In the case of $S_n(^{14}C)$, since there has been no direct measurement[1] of a doublet mass

TABLE IV. Neutron binding energies obtained from mass differences (Refs. 1, 7) and the present $S_n(^2H)$ value. Also shown are the calculated energies of the capture state-to-ground state γ rays emitted following slow neutron capture into those nuclei.

(n,γ) Reaction product nucleus	$S_n(^{A+1}Z)$ (keV)	Nucleus recoil energy (keV)	$S_n(^{A+1}Z) \rightarrow$ g.s. γ-ray energy (keV)	Error (eV) σ_m	σ_γ	σ_t
^{3}T	6257.268	6.953	6250.316	17	16.5	24
^{13}C	4946.329	1.010	4945.319	20	12.9	24
^{14}C	8176.483	2.561	8173.922	33	21.9	40
^{15}N	10833.297	4.196	10829.101	24	29.3	38

difference involving $^{13}C - ^{14}C$, we obtained a value of 7937.883 ± 0.031 μu for the $^{13}C^{1}H - ^{14}C$ mass difference based on the adjusted values of the mass excesses[1]; i.e., from the summation $[(^{13}C - 13) + (^{1}H - 1) - (^{14}C - 14)]$.

Since each of the lower mass ^{A}Z isotopes used in the determination of the $S_n(^{A+1}Z)$ listed in Table IV are stable, these same ^{A+1}Z nuclei can also be produced in neutron capture reactions with the ^{A}Z stable-element targets. Furthermore, the product nucleus following slow neutron capture is at an excitation energy equal to $S_n(^{A+1}Z)$. Thus in these few cases, equally precise energies can be computed for the γ rays emitted in the direct capture state-to-ground-state transitions by applying the appropriate corrections for nuclear recoil (shown in column 3 of Table IV). These primary γ-ray energies are also included in Table IV. Gamma-ray energies determined in this manner have, in recent years, provided the most accurate calibration standards available in neutron capture γ-ray spectroscopy[5,6] in the energy region above ~3.5 MeV, and have therefore proven to be of great practical importance to provide such energy calibrations.

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