Two-quantum radiative thermal neutron capture in ^{1}H

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A value of (-5.2 ± 6.4) μb has been obtained for the cross section of the two-quantum radiative thermal neutron capture in hydrogen for the energy region $330 < E_{\gamma 1}, E_{\gamma 2} < 1890$ keV. Restriction of the evaluation to a γ energy range from 700 to 1520 keV yields (-0.4 ± 3.0) μb. The present results are weighted averages from several coincidence experiments performed with various types of detectors. In one experiment, using two NaI(Tl) detectors, special care was taken to cover also the region of lower γ-ray energies ($233 < E_{\gamma 1}, E_{\gamma 2} < 2017$ keV), since here the most severe discrepancy to theoretical predictions is indicated by an earlier measurement. The existing data reject a hypothesis concerning nonorthogonality of the $^3S_1$ neutron-proton capturing and the deuteron ground state. Due to use of a H_2O target from the present measurements at the same time the total cross section for the reaction $^{16}O(n,\gamma)^{17}O$ could be determined as (187 ± 10) μb.

[NUCLEAR REACTIONS $^1H(n,\gamma\gamma)$, $^{16}O(n,\gamma)$, thermal n, measured $\sigma_{\gamma\gamma}$, σ_γ.]

I. INTRODUCTION

In the last few years the process of two-photon emission following thermal neutron capture in hydrogen has drawn much attention because of the possibility of deriving additional information on the neutron-proton interaction. Two-photon emission itself has already been studied in atomic transitions leading to excellent agreement with theory.[1]

A long existing 8% discrepancy between calculation and measurement of the $^1H(n,\gamma)$ cross section seemed to hint at some principal failure of the conventional assumptions about the two-nucleon problem. Breit and Rustgi[2] assumed that part of the reaction could go via the $^3S_1$ capturing state which normally is believed to be orthogonal to the deuteron ground state, so that the $M1$ transition probability should vanish. On the premises of nonorthogonality being responsible for the missing 8% of the one-photon cross section, Adler[3] calculated a two-photon cross section as high as 42 μb. In this case the two-photon process proceeds via the A^2 or gauge term which is proportional to the overlap of the np capturing state and the deuteron ground state. A recent reevaluation by Blomqvist and Ericson[4] yields a value of 20 μb instead of 42 μb. The hypothesis of nonorthogonality, however, is no longer necessary to explain the experimental $^1H(n,\gamma)$ cross section. Riska and Brown[5] and Gari and Huffmann[6] have shown independently that the proper inclusion of meson exchange currents yields an additional contribution to the theoretical cross section so that the experimental value is reproduced by the calculation with an accuracy of about 1%. Further, two recent papers by Riska[7] and Friar[8] conclude that energy dependence of the Hamiltonian does not change orthogonality between eigenstates with different energies, so that nonorthogonality is a very unlikely postulate.

Normally, the $^1H(n,\gamma\gamma)^2H$ process is thought to be due to the dispersive term which involves intermediate states. For its cross section a value of 0.12 μb has been predicted independently by several theoretical groups.[4,9-11]

In contrast to the very small value of 0.12 μb a two-photon cross section of 42 μb and even of 20 μb is within experimental reach. An upper limit of 1000 μb was obtained for $\sigma_{\gamma\gamma}$ in a first measurement.[12] Two years later, a two-photon capture cross section of (350 ± 53) μb was reported[13] for the γ-energy range from 600 to 1620 keV. This result, remarkably exceeding Adler's upper limit, could not even be explained by the theorists.[4,10,14] The unexpected result[13] is probably caused by several effects[15,16] by which photons from the reaction $^1H(n,\gamma)^2H$ induce a response of both detectors in the coincidence measurement. Shortly afterwards, Earle *et al.*[17] and Wüst *et al.*[18] independently carried out experiments that yielded results of $\sigma_{\gamma\gamma} < 33$ μb (Ref. 17) and $\sigma_{\gamma\gamma} = (-28 \pm 49)$ μb (Ref. 18) for the energy range 600 keV$<E_{\gamma 1}$, $E_{\gamma 2}<$1620 keV, and 46 keV$<E_{\gamma 1}$, $E_{\gamma 2}<$2177 keV, respectively. Both results are inconsistent with the previous experimental result.[13] In a later measurement Earle *et al.*[19] obtained an improved value of (-3 ± 8) μb for the energy range from 600 to 1620 keV. This result rules out Adler's value[3] of 27 μb (for $E_\gamma = 600-1620$ keV) based on the assumption of "maximal nonorthogonality," defined[3] as making the overlap $\langle ^3S_d | ^3S_{np} \rangle$ of the triplet states at different energies equal to the overlap $\langle ^3S_d | ^1S_{np} \rangle$ of the triplet and singlet states. Another theoretical value of 13 μb for the same energy range, recalculated recently by Blomqvist *et al.*[4] assuming the same nonorthogonality conditions, still was within the range of less than two standard deviations of the Chalk River value.[19] Also a partial violation of orthogonality could be thought of, which would result in a total two-quantum cross section somewhere between 20 and 0.12 μb for $0<E_\gamma<2.22$ MeV. This situation made desirable a more accurate investigation of the reaction $^1\mathrm{H}(n,\gamma\gamma)^2\mathrm{H}$. Meanwhile, Earle and McDonald,[20] using two big NaI(Tl) detectors, succeeded in deducing an upper limit of $\sigma_{\gamma\gamma}=1.6$ μb.

This result, however, only covers the energy interval 700 keV$<E_{\gamma 1}, E_{\gamma 2}<$1520 keV. An earlier measurement[13] indicates an increase in the differential cross section $d\sigma_{\gamma\gamma}/dE_\gamma$ towards lower γ-ray energies. It has been suggested[15,16] that crosstalk may well account for both the yield and the shape of the original coincidence spectrum. To prove this suggestion and to reduce the statistical error published earlier,[18] in one of the present measurements both γ-energy thresholds were set below the maximum of the crosstalk energy spectrum which is awaited[19] at $E_{\gamma 1}$ or $E_{\gamma 2}\sim 250$ keV.

II. EXPERIMENTAL PROCEDURE

The method suitable for registering the two simultaneously emitted, continuously distributed γ rays is a coincidence experiment with two γ-ray detectors. Since accidental coincidences originating from the Compton tails of the 2.2 MeV one-photon transitions are a principal limiting factor in this measurement, the detectors should have a high peak-to-total ratio (see Fig. 1), and their full-energy peak efficiencies as well as energy and time resolutions should be optimal. As these requirements cannot simultaneously be met by a single type of detector, it is preferable to carry out several runs using different detector combinations.

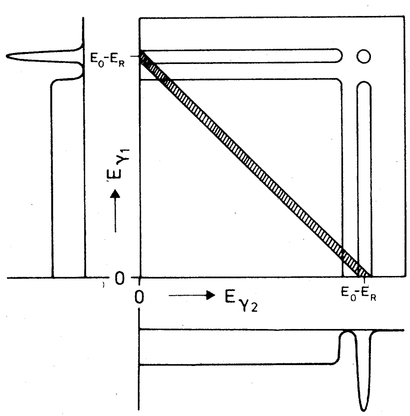

FIG. 1. Schematic representation of the coincidence matrix and the band in which full energy events of the true γ-γ coincidences are expected. $E_0=2224.7$ keV is the deuteron binding energy which has to be corrected for the recoil energy E_R.

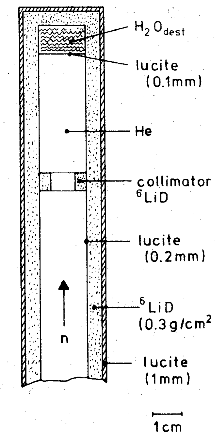

FIG. 2. Target geometry and shielding used in the present experiments.

A serious complication in the coincidence experiments is the γ-ray cross registration mentioned above which can simulate real two-photon events. This effect in connection with the two-photon experiment has already been studied in detail elsewhere.[15,16,19] The scattering of γ radiation from one detector into the other has to be avoided by proper shielding which severely restricts the geometrical arrangement.

Our experiments were performed at the external neutron beam[21] of the FRJ-2 (DIDO) research reactor of the KFA Jülich. For low background the beam is filtered by a 20 cm long Bi single crystal. The neutron flux is about 2×10^8 cm^{-2} s^{-1} at the target position. Details of the target arrangement are depicted in Fig. 2. For the reduction of γ-ray cross registration in our experiments, a wedge of lead[18] or a block of lead (see Fig. 3) was placed between the detectors.

The main contribution to crosstalk derives from

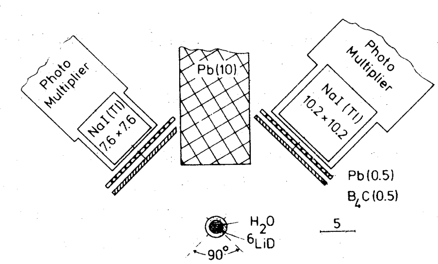

FIG. 3. Arrangement of the detectors and shielding against crosstalk in the present experiment (dimensions given in cm).

single Compton scattering and pair production, involving photons with energies of ~260 keV in our geometry and 511 keV, respectively. To give an upper limit for the crosstalk one has to estimate the ratio of quanta emitted from one detector and detected in the other over the number of 2.2 MeV quanta detected in the first one and then to regard the attenuation by the shielding. In calculating the attenuation one has to allow for[22] the contribution from γ rays which are inelastically scattered within the shield but only lose an amount of energy comparable to the energy resolution [~90 keV in the NaI(Tl), ~4 keV in the Ge(Li) measurements]. For the NaI(Tl) measurement we calculate an upper limit of 2.6×10^{-3} μb for a two-photon cross section simulated by this low-energy crosstalk, whereas in the Ge(Li) measurements the upper limit is only 6.5×10^{-5} μb.

Lee and Earle[16] have calculated the contribution from multiple Compton scattering and positron annihilation in flight, both processes being responsible for higher energy crosstalk. For a detector geometry very similar to ours these effects simulate 1×10^{-5} two-photon events per single-photon event without any shielding. This high energy crosstalk is caused by scattered γ quanta with most probable energies around 800 keV.[16] At 1 MeV γ-ray energy the transmission through our lead shield, again including[22] the γ rays degraded in energy by only a small amount, is 1.7×10^{-3} for the NaI(Tl) and 1×10^{-4} for the Ge(Li) measurements. By use of these values we can estimate a high-energy contribution to crosstalk of about 6×10^{-3} μb and 2×10^{-4} μb for the NaI(Tl) and the Ge(Li) measurements, respectively.

The numbers quoted above show that by our shielding arrangement crosstalk is suppressed sufficiently in regard to both the cross section expected theoretically and the experimental error given below. For the reduction of systematic errors, three different pairs of detectors were used. The first measurement with two medium-size Ge(Li) detectors has already been published.[18] A second experimental arrangement with two NaI(Tl) scintillators (10.2 cm×10.2 cm and 7.6 cm×7.6 cm) is shown in Fig. 3. For a third and fourth run we used two big Ge(Li) detectors of 120 and 127 cm^3 with relative efficiencies of 19% and 24% in a geometry similar to the one shown in Fig. 3.

The methods of data sampling and analysis were nearly the same for all experiments. Coincidences were registered by a conventional fast-slow system[23] and recorded on magnetic tape event by event. The data were analyzed off-line at our central IBM/I370-168 computer. From the coincidence data a two-dimensional representation was

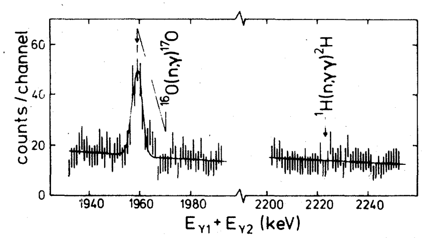

FIG. 4. Partial sum spectrum from the third Ge(Li) measurement, energy range E_{γ_1} = 1000–1111 keV. The full lines give the results of the fits.

generated as schematically shown in Fig. 1. In this coincidence array all events whose energies $E_{\gamma1}$ and $E_{\gamma2}$ sum up to the same energy, such as the $^1H(n,\gamma\gamma)$ events do, lie on a diagonal strip. By summing all events that belong to a given sum energy, a one-dimensional spectrum is obtained. In order to take into account the energy dependence of the coincidence efficiency the coincidence array

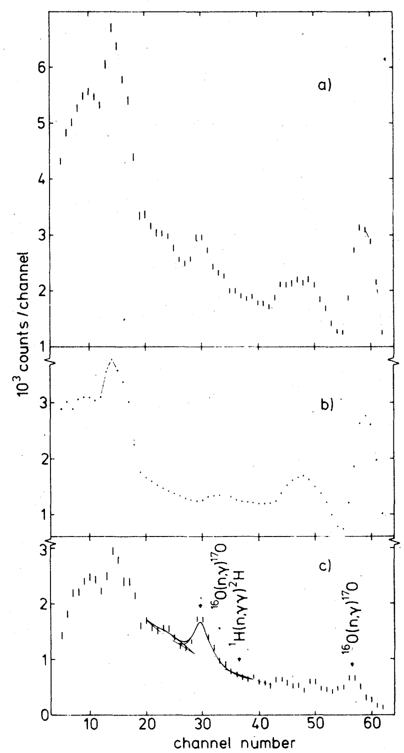

FIG. 5. Partial sum spectra (γ-ray sum energy 40.6 keV/channel) from the NaI(Tl) measurement, energy range E_{γ_1} = 828–1026 keV [(a) is original coincidence spectrum, (b) is the normalized singles spectrum (statistical errors negligible), and (c) is the spectrum of real coincidences, obtained by subtracting (b) from (a). The full lines give the result of the fit]. The arrows labeled with the $^{16}O(n,\gamma)^{17}O$ reaction indicate the peaks which correspond to the 1088–(2184)–871 keV and 2184–871 keV coincidences (sum energy 1959 and 3055 keV, respectively).

TABLE I. Results of the NaI(Tl) coincidence measurement.

$\Delta E_{\gamma 1}$ (keV)	N [a]	ΔN_{stat} [b] (counts)	ΔN_{bg} [c]	ΔN_{tot}	$\sigma_{\gamma\gamma}$ [d] (μb)
233–432	0	124	42	131	0.0 ± 8.3
432–630	–49	151	52	160	-1.1 ± 3.6
630–828	–132	101	37	108	-2.4 ± 1.9
828–1026	–93	102	38	109	-1.6 ± 1.9
1026–1224	–16	101	37	108	-0.3 ± 1.9
1224–1424	–10	105	42	113	-0.2 ± 2.0
1424–1621	–112	102	34	108	-1.9 ± 1.9
1621–1819	176	154	52	163	3.5 ± 3.2
1819–2017	86	136	59	148	3.6 ± 6.1

[a] Number of counts in the peak as determined from the fit.

[b] Statistical error $= (\bar{n}_p \cdot p)^{1/2}$, where $\bar{n}_p$ is the average number of counts per channel in the peak region of p channels.

[c] Error of background in the peak region $= (p/b)\ (b\bar{n}_b)^{1/2}$ where $\bar{n}_b$ is the average number of counts per channel in the background-fit region of b channels.

[d] Calculated with use of the formula given in the text.

was divided into strips of width $\Delta E_{\gamma 1} = 200$ keV for the NaI(Tl) measurement and $\Delta E_{\gamma 1} = 111$ keV for the Ge(Li) measurements. For each of the strips a sum spectrum was established. Figure 4 shows one of these spectra from one of the Ge(Li) measurements while Fig. 5(a) shows a similar spectrum from the NaI(Tl) measurement. As can be seen from Fig. 4, the background in the Ge(Li) sum spectra is smooth and can easily be determined by averaging over a region large in comparison to the peak region, so that the background error is negligible. In the NaI(Tl) measurement, however, the high contribution of random coincidences makes background determination more difficult [Fig. 5(a)]. By subtracting a singles spectrum normalized with the intensity of the random coincidences in the 2.2 MeV full energy peak [Fig. 5(b)] the random coincidences are eliminated and a sum spectrum of purely real coincidences [Fig. 5(c)] is obtained in which the background is smoother. It can be fitted by use of an exponential function on which Gaussian-shaped peaks are superposed. Since in the NaI(Tl) spectra the number of data points which can be used for background determination is not so large compared to the peak region, the background errors must be taken into account (Table I). For all measurements the background is determined first, and then the area of the two-quantum peak is fitted with background, peak position, and peak shape fixed, the latter having been determined from the oxygen coincidence peaks (see below).

The relation between the intensity N obtained through the fit and the two-photon cross section for the corresponding γ-energy range is given by the following equation:

$$\sigma_{\gamma\gamma} = \frac{N\sigma_\gamma}{[\epsilon^{(1)}(E_\gamma)\,\epsilon^{(2)}(2.2\text{ MeV} - E_\gamma) + \epsilon^{(1)}(2.2\text{ MeV} - E_\gamma)\,\epsilon^{(2)}(E_\gamma)]\,\Omega_1\Omega_2 A T}\,.$$

Here σ_γ denotes the ^{1}H capture cross section (334.2 ± 0.5 mb[24]), T the measuring time, A the measured single-photon emission rate, Ω_i the solid angle subtended by detector i, and $\epsilon^{(i)}$ the absolute coincidence full-energy peak efficiency of detector branch i. Figures 6(a) and 6(b) show the differential $^1\mathrm{H}(n, \gamma_1\gamma_2)$ cross section obtained in the NaI(Tl) and in one of the Ge(Li) measurements.

The two spectra of Figs. 4 and 5 contain a prominent coincidence peak at the sum energy $E_{\gamma 1} + E_{\gamma 2} = 1959$ keV. It is due to the coincident 871 keV and 1088 keV γ rays emitted as an $(82 \pm 3)\%$ branch[25] after thermal neutron capture in ^{16}O of the water. Two earlier measurements of the total $^{16}\mathrm{O}(n, \gamma)\,^{17}\mathrm{O}$ cross section have been reported yielding (178 ± 25) μb (Ref. 26) and (202 ± 27) μb.[25] From our data (Table II) we derive a value of (153 ± 6) μb for the partial cross section of the 1088–(2184)–871 keV cascade which with the branching ratio given above results in (187 ± 10) μb for the total capture cross section. The good agreement with the earlier values confirms our calibration of absolute coindidence efficiency which was obtained with calibrated sources of ^{152}Eu, ^{22}Na, ^{60}Co, and ^{88}Y at the target position.

III. RESULTS AND CONCLUSION

The individual results for the two-photon cross section from the different runs are shown in Table II. In all experiments the energy region from 330

TABLE II. Summary of the results of the present measurements on the two-quantum radiative thermal neutron capture cross section $\sigma_{\gamma\gamma}$ of ^{1}H and the partial thermal neutron capture cross section σ'_γ of ^{16}O [1088-(2184)-871 keV γ cascade].

Detectors	Time resolution[a] (*ns*)	Measuring time (*d*)	^{1}H: $\sigma_{\gamma\gamma}$ (μb)[c] 330–1890 keV	^{1}H: $\sigma_{\gamma\gamma}$ (μb)[c] 700–1520 keV	^{16}O: σ'_γ (μb)[c]
Ge(Li) 58 cm^3/Ge(Li) 62 cm^3	23	34	5 ± 32 [b]		171 ± 22
NaI(Tl) 7.6 × 7.6 cm/NaI(Tl) 10.2 × 10.2 cm	4	35	−2.7 ± 9.5	−4.8 ± 4.0	152 ± 8
Ge(Li) 120 cm^3/Ge(Li) 127 cm^3	9	30	−3 ± 16	10.6 ± 8.6	160 ± 16
Ge(Li) 120 cm^3/Ge(Li) 127 cm^3	11	49	−10.6 ± 10.8	3.3 ± 5.3	140 ± 18
Weighted averages			−5.2 ± 6.4	−0.4 ± 3.0	153 ± 6

[a] FWHM obtained with ^{22}Na source. During the measurements, events were accepted in a time window of 1.2 × FWHM. In the last run with the big Ge(Li) detectors lower energy thresholds were used which results in a slightly worse time resolution.

[b] Experiment already reported (Ref. 18), improved value from reevaluation.

[c] The given errors include those from the coincidence peak determination and errors of the absolute coincidence efficiencies.

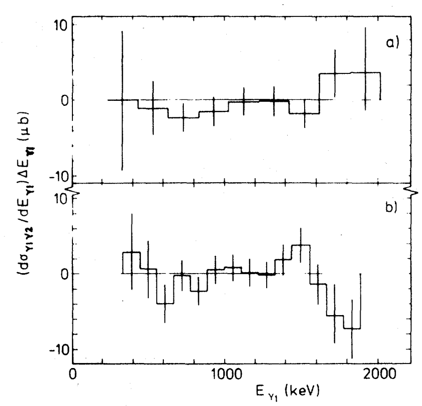

FIG. 6. Differential cross section for the ^{1}H$(n,\gamma\gamma)^2$H reaction [(a) is from the NaI(Tl) measurement, (b) is from the second measurement with the big Ge(Li) detectors].

to 1890 keV was observed. As all four measurements have been carried out independently, the weighted average of the four numbers—$\sigma_{\gamma\gamma} = (-5.2 \pm 6.4)$ μb—is the final result of this work. The error is the single standard deviation. The result of this work is in agreement with the value of $\sigma_{\gamma\gamma} < 1.6$ μb obtained by Earle *et al.*[20] For comparing the two results one should mention that the energy region covered by Earle *et al.*[20] is from 700 to 1520 keV only. If the evaluation of our measurements is restricted to the same energy interval (see Table II) a weighted average of (-0.4 ± 3.0) μb results for the two-photon partial cross section. The results of the present NaI(Tl) measurement [$\sigma_{\gamma\gamma} = (0.4 \pm 12.1)$ μb for 233 keV $< E_{\gamma 1}, E_{\gamma 2} <$ 2017 keV, Table I] and of our earlier Ge(Li) measurement[18] show that with appropriate shielding no contribution to $\sigma_{\gamma\gamma}$ exceeding the statistical error is obtained even in a γ-energy interval in which the crosstalk spectrum reaches maximum.[19] They disprove the energy dependence of the differential ^{1}H$(n,\gamma\gamma)$ cross section given earlier[13] which would correspond to what is awaited for two-photon cascades of the type $M1-M1$.[4] If for the differential cross section the energy dependence of the $E1-E1$ mode[4] is supposed, an upper limit of $\sigma_{\gamma\gamma}(E1-E1) < 3.2$ μb for the total two-photon ^{1}H capture cross section follows from $\sigma_{\gamma\gamma} < 1.6$ μb (Ref. 20) (700 keV $< E_{\gamma 1}, E_{\gamma 2} <$ 1520 keV). The experimental accuracy is still short by orders of magnitude to verify the very precise prediction of (0.1176 ± 0.0003) μb (Ref. 5) from model-independent calculations based on conventional assumptions.

On the basis of the experimental upper limit $\sigma_{\gamma\gamma} = 3.2$ μb, the absolute square of the overlap integral for the $^3S_{np}$ and the ^3S_d states is smaller than 16% of the overlap of the $^1S_{np}$ with the deuteron ground state, at a significance level of 68% (single standard deviation). Although this value rejects the idea of a "maximal nonorthogonality" as defined by Adler, which should lead to a 2-photon cross section of 20 μb,[4] it still would leave room for speculation on a severe violation of the orthogonality rule. Yet a much more definitive upper limit for the deviation from orthogonality can be concluded from a very accurate measurement of the circular polarization of the 2.2 MeV ^{1}H(n,γ) radiation done by Kolomensky *et al.*[27] This measurement yields an upper limit of 2×10^{-4} for the ratio of triplet to singlet capture. With use of the formula given in Refs. 2, 27, and 28 from this value an upper limit of 1.4×10^{-3} can be deduced for the ratio $|\langle{}^3S_d|{}^3S_{n,p}\rangle/\langle{}^3S_d|{}^1S_{n,p}\rangle|^2$.

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