# Upper limit for the ${}^{1}H(n, \gamma\gamma){}^{2}H$ cross section

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A value of  $(-0.8 \pm 2.5) \times 10^{-5}$  has been measured for the ratio of double-photon to single-photon emission following neutron capture in hydrogen for 600 keV  $< E_{\gamma} < 1620$  keV. The two Ge(Li) detectors used in the experiment subtended an angle of 85° at the H<sub>2</sub>O target and were shielded from each other to reduce the background from the cross registration of single  $\gamma$  rays. The upper limit of the measured two-photon cross section  $(-3 \pm 8 \,\mu b)$  is two orders of magnitude larger than the most recent theoretical predictions ( $\approx 0.07 \,\mu b$ ) for this energy range.

NUCLEAR REACTIONS <sup>1</sup>H $(n, \gamma\gamma)$ , thermal *n*, measured  $\sigma_{2\gamma}$ , Ge(Li) detectors.

# I. INTRODUCTION

The  ${}^{1}H(n, \gamma){}^{2}H$  and the competing  ${}^{1}H(n, \gamma\gamma){}^{2}H$  reactions are of particular theoretical interest since the cross sections for these simple processes can be calculated with few assumptions. Any significant difference between experiment and theory for these two cross sections would seriously undermine current theories of quantum electrodynamics. Two-photon emission has been searched for and found for transitions in other nuclei, particularly<sup>1-3</sup> from the first 0<sup>+</sup> states of  ${}^{16}O$ ,  ${}^{40}Ca$ , and  ${}^{90}Zr$ where the branching ratios  $(\Gamma_{_{2\gamma}}/\Gamma_{_{1\gamma}}\approx\!3\!\times\!10^{-4})$  have been measured. Compared to the  ${}^{1}H(n, \gamma\gamma)^{2}H$  reaction the double-photon cross sections for these nuclei are easier to measure since the competing single-photon transition  $(0^+ \rightarrow 0^+)$  is forbidden. However, calculations for these cases involve details of nuclear structure as well as quantum electrodynamics.

For many years an 8% discrepancy existed between the experimental and theoretical  ${}^{1}H(n, \gamma)^{2}H$ cross sections for thermal neutrons. It required the introduction<sup>4-7</sup> into the calculations of one pion exchange terms and of  ${}^{3}D_{1}$  terms in the deuteron ground state to remove this discrepancy. An alternative suggestion by Breit and Rustgi,<sup>8</sup> that the discrepancy might be resolved by assuming that the  ${}^{3}S_{1}$  np capturing state and the deutron ground state were not orthogonal, prompted Adler<sup>9</sup> to calculate that the two-photon cross section would be  $\approx 42 \ \mu$ b under the assumption of sufficient nonorthogonality to explain the 8% discrepancy in the single-photon cross section. It has been recently argued, however,<sup>10,11</sup> that this nonorthogonality hypothesis is untenable.

Following Adler's calculation,<sup>9</sup> Arnold *et al.*<sup>12</sup> reported an upper limit of 700 µb for the two-photon cross section. Dress *et al.*<sup>13</sup> subsequently reported a cross section ( $\sigma_{2\gamma} = 350 \pm 50 \mu$ b) for  $\gamma$  rays in the energy range 600 keV  $< E_{\gamma} < 1620$  keV. This latter measurement, which was a factor of 10 larger than Adler's calculated cross section and more than  $3 \times 10^4$  times larger than the cross section calculated by Grechukhin<sup>14</sup> assuming conventional wave function orthogonality, stimulated considerable theoretical<sup>15-21</sup> and experimental<sup>22-24</sup> work on the determination of the  ${}^{1}\text{H}(n, \gamma\gamma){}^{2}\text{H}$  cross section.

Independent measurements by Earle et al.<sup>22,23</sup> and Wüst et al.<sup>24</sup> have set a much lower limit for this cross section. Also Alburger<sup>25</sup> and Lee and Earle<sup>26</sup> showed that photons from the  ${}^{1}H(n, \gamma)^{2}H$  reaction scattering from one detector into the other (cross registration) may have been an important factor in the earlier measurement.  $^{13}\,$  We have further improved the sensitivity and have determined an upper limit for the  ${}^{1}H(n, \gamma\gamma)^{2}H$  cross section ( $\sigma_{2\gamma} < 8 \ \mu b$ ) for 600 keV  $< E_{\gamma} < 1620$  keV. For the E1-E1 cascade the two-photon cross section for the energy range 600 keV  $\leq E_{\gamma} \leq 1620$  keV is ~60% of the two-photon cross section for 0 keV  $< E_{\gamma} < 2223$  keV. The observed limit is lower than the most recently calculated<sup>15</sup> cross section  $(\sigma_{2\gamma} \approx 13 \ \mu b)$  for the range 600 keV <  $E_{\gamma}$  < 1620 keV obtained by assuming that the  ${}^{3}S$  capture state and the deuteron wave functions are nonorthogonal, but it is larger than the result of more conventional calculations<sup>15,19</sup>  $\sigma_{2\gamma} = 0.07 \ \mu b$ .

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#### **II. EXPERIMENTAL DETAILS**

#### A. Equipment

The present experiment was performed with 0.009 eV neutrons obtained by Bragg reflecting a beam of neutrons from the Chalk River NRU reactor thermal column with a pyrolitic graphite monochromator. The scattering cross section for <sup>1</sup>H is 100 times the capture cross section, and so this neutron beam is effectively thermalized to 0.025 eV in the target before capture. The beam traveled down a 4 cm inside diameter tube lined with 5 mm of enriched <sup>6</sup>LiF (>90% <sup>6</sup>Li) to a 50 cm<sup>3</sup> distilled H<sub>2</sub>O sample contained in a bag made of 0.1 mm thick polyethylene (Fig. 1). The <sup>6</sup>LiF shield provided a neutron attenuation factor of greater than  $10^{14}$ , effectively shielding the detectors from neutrons in the beam and those scattered from the target. The neutron flux at the target was  $4.4 \times 10^5$  neutrons s<sup>-1</sup> cm<sup>-2</sup>. Two Ge(Li) detectors having photopeak efficiencies of 11.3% and 13.3% at 1.33 MeV (relative to a 7.6 cm  $\times$  7.6 cm NaI detector at 25 cm) were placed as close to the target as possible (Fig. 1) and were shielded from each other by 4.9 cm of heavy metal (composition 90% W, 6% Ni, 4% Cu,  $\rho = 16.7 \text{ g cm}^{-2}$ ). In addition the target-detector assembly was surrounded by 10 cm of Pb. The total counting rate in each Ge(Li) detector was 4.5  $\times 10^{3} \text{ s}^{-1}$ .

The linear signals from the two detectors were summed and gated by pulses from a fast-slow coincidence circuit before analysis by an analog to digital converter. The fast coincidence time resolution was <6 ns [full width at half maximum (FWHM)] and was achieved by using two ORTEC 473 constant fraction discriminators (CFD) in a



FIG. 1. The target-detector configuration used in the present experiment. The Pb shield, shown schematically, surrounded the target, coaxial Ge(Li) detectors, and flight tube which are shown to scale.

slow-rise-time-rejection mode. This mode rejected low energy pulses; 50% of the pulses at 350 keV and 20% at 500 keV were rejected by the CFD's. The slow coincidence circuit vetoed all coincidence events unless the  $\gamma$ -ray energy deposited in each detector was between 600 and 1620 keV. The neutron fluence was monitored by recording the yield of 2.223 MeV  $\gamma$  rays from the <sup>1</sup>H $(n, \gamma)^2$ H reaction.

For one set of measurements a PDP-5 computer was used on line to store on magnetic tape the  $\gamma$ ray pulse height from each detector and the fast timing information as determined by the time to amplitude converter. This related address information was subsequently analyzed with the aid of a PDP-10 computer.

#### B. Gamma-ray cross registration

An important experimental complication in low cross section measurements of coincident  $\gamma$  rays in the presence of an intense  $\gamma$ -ray flux is detector cross registration. In the present experiment coincidence events caused by the scattering of a single 2.223 MeV  $\gamma$  ray from one detector into the other such that the total energy deposited in the two detectors is 2.223 MeV are indistinguishable from true  ${}^{1}\text{H}(n, \gamma\gamma){}^{2}\text{H}$  events. They must be reduced by shielding between the detectors, and any residual contribution must be well defined to avoid confusion with true  ${}^{1}\text{H}(n, \gamma\gamma){}^{2}\text{H}$  events.

The extent of this problem is exhibited in Fig. 2, which shows the coincidence sum spectrum near 2.2 MeV with and without 4.9 cm of heavy metal between the detectors. These spectra have been normalized to the same neutron fluence. In addition to reducing the smooth background around 2.223 MeV, the heavy metal greatly reduces the coincidence events summing to 2.223 MeV. Since the heavy metal does not significantly attenuate  $\gamma$  rays reaching the detectors directly from the H<sub>2</sub>O target (Fig. 1) but does attenuate those crossing between the detectors, it is clear that the majority of the events observed at 2.223 MeV without the shielding are due to cross registration.

The two main processes responsible for cross registration are Compton scattering and pair production. Figure 3 illustrates a spectrum of events observed in detector A with the restriction that  $E_1 + E_2 = 2.223 \pm 0.004$  MeV. This spectrum was obtained by sorting three parameter events recorded by the PDP-5 computer with the target-detector geometry as shown in Fig. 1, but with the heavy metal removed. The large peak at about 300 keV is primarily caused by events in which the incident 2.223 MeV  $\gamma$  ray Compton scatters from detector B into detector A. The peak at 511 keV results



FIG. 2. Sum spectra of coincidences summing to energy  $E_1+E_2$  obtained with two Ge(Li) detectors in the geometry of Fig. 1. Crosses indicate the spectrum obtained without the 4.9 cm thick heavy metal shield and the points indicate the spectrum with the shield in place. The latter spectrum was normalized to the same neutron fluence as the former spectrum.

from pair production in detector B and reabsorption of one 511 keV photon by each detector. The spectrum is approximately symmetric about 1.11 MeV since the detectors are similar in size and the first interaction of the 2.223 MeV  $\gamma$  ray can be in either detector. Because of the large increase in yield below 600 keV and above 1620 keV it was decided to accept only events between these energies and to use shielding to reduce the cross registration events in this region.

Cross registration above 600 keV has been discussed by Alburger<sup>25</sup> and by Lee and Earle.<sup>26</sup> The calculations of Lee and Earle indicate that two higher order forms of cross registration can be significant in this energy region; multiple Compton scattering (MCS) and  $\gamma$  rays from annihilation in flight of positrons from pair production in one of the detectors (APF).

It was assumed in these calculations that all 2.223 MeV  $\gamma$  rays contributing to cross registration originated from the H<sub>2</sub>O target. In the present experiment the 10 cm Pb shielding reduced the contribution from 2.223 MeV  $\gamma$  rays originating outside the H<sub>2</sub>O target to less than 10<sup>-4</sup> of the flux of 2.223 MeV  $\gamma$  rays from the target. Tests of

cross registration for  $\gamma$  rays originating directly behind one of the detectors were made with the 1.78 MeV  $\gamma$  rays from <sup>28</sup>Al. These tests showed that the 2.223 MeV room background gave a negligible number of coincidence events.

Monte Carlo calculations<sup>26</sup> show that the number of MCS events increases rapidly as the scattering angle decreases. There is a contribution in the energy region near 600 keV for the present geometry where the centers of the two Ge(Li) detectors subtend an angle of  $85^{\circ}$  at the center of the H<sub>2</sub>O target. However, MCS is negligible when the detectors are on opposite sides of the target as they were in the experiment reported by Dress *et al.*<sup>13</sup>

On the other hand, APF contributes significantly over the whole energy range regardless of the target-detector configuration. The calculations<sup>26</sup> indicate that the majority of the events observed for 600 keV  $\langle E_{\gamma} \rangle$  1620 keV in the present geometry arise from APF. These calculations, which predict (88 ± 22)% of the events measured [Fig. 2(A)], also indicate that the spectra of events from APF and MCS decrease monotonically as the energy of the crossover  $\gamma$  ray increases. This conclusion is confirmed by the shape of the spectrum shown in Fig. 3.



FIG. 3. The spectrum in one detector of  $\gamma$  rays in coincidence with  $\gamma$  rays in the other detector with the restriction that  $E_1 + E_2 = 2.223 \pm 0.004$  MeV. A background spectrum measured with  $E_1 + E_2 = 2.235$  MeV has been subtracted.

Since the detailed spectral shape for events due to cross registration is a complicated function of several poorly known parameters, it was decided to estimate the residual cross registration with shielding by using the measured spectrum shown in Fig. 3. The spectrum of  $\gamma$  rays striking the heavy metal was unfolded from Fig. 3 by assuming that the yield decreased linearly from 1.11 MeV to zero at 1.72 MeV. The attenuation of the 4.9 cm of heavy metal was measured with radioactive sources and interpolated using tabulated<sup>27</sup>  $\gamma$ -ray linear attenuation coefficients. This method of determining the number of residual cross-registration events was corroborated in a measurement with 2.5 cm of Pb shielding<sup>22</sup> where the measured attenuation agreed with the calculated value. An overall uncertainty of  $\sim 20\%$  was estimated for the calculated value of the residual cross-registration events with the heavy metal shielding. This uncertainty is large enough to include the possibility that the spectrum of cross-registration  $\gamma$  rays striking the heavy metal is constant from 1.11 to 1.72 MeV.

### C. Other background

The remaining limitation on the sensitivity of the experiment to real two-photon events arises from the flat background shown by solid points in Fig. 2(B). The dominant source of this background, contributing more than 60% of the events detected at 2.223 MeV, was from random summing of the Compton tails of two 2.223 MeV  $\gamma$  rays from the

<sup>1</sup>H $(n, \gamma)^2$ H reaction. This contribution was kept low by making the time and energy resolution as good as possible.

The remainder of the flat background is from real coincidences of neutron capture  $\gamma$  rays from materials other than <sup>1</sup>H which deposit a fraction of their energy in the two detectors such that  $E_1 + E_2$ = 2.223 MeV. There is no contribution from <sup>6</sup>Li capture in the <sup>6</sup>LiF lining of the beam tube because there are no two step  $\gamma$ -ray cascades in <sup>7</sup>Li having both  $\gamma$  rays greater than 600 keV. There is a significant contribution from <sup>20</sup>F, although a large fraction of its cascades have only one  $\gamma$  ray greater than 600 keV. A beam tube of <sup>6</sup>Li<sup>2</sup>H would have been preferable but was unavailable. The <sup>15</sup>N  $\gamma$ ray background from air in the tube was made negligible by flushing He gas through the tube. Finally neutron capture in  $^{16}\text{O},\,\sigma\approx190\,\mu\text{b}$  (see Sec. II D) contributed but, as is shown in the next section, the  $^{16}O(n, \gamma)^{17}O$  reaction also provided an important calibration for the  ${}^{1}H(n, \gamma\gamma)^{2}H$  measurement.

# D. $\sigma_{2\gamma}$ measurement

The  ${}^{1}\text{H}(n, \gamma\gamma)^{2}\text{H}$  sum spectrum from 1.9 to 2.3 MeV is shown in Fig. 4. The electronic conditions for an acceptable event were that the two  $\gamma$  rays were in fast coincidence ( $\Delta t = 16$  ns) and that the energy deposited in each detector was between 600 and 1620 keV. The data plotted in Fig. 4 are the sum of 25 runs, each of about 24 h duration,



FIG. 4. A portion of the sum spectrum recorded in the geometry of Fig. 1. The accumulated counts from 25 runs are shown. The peak at 1.959 MeV results from the summation of 0.871 and 1.088 MeV coincident  $\gamma$  rays which follow neutron capture in <sup>16</sup>O.

and were collected over a period of 45 days. The maximum linear gain shift was 2 keV. Thus some spectra had to be shifted by one channel before summation. No other corrections were applied to the data.

The peak in Fig. 4 at 1.959 MeV is due to the summation of the 0.871 and 1.088 MeV cascade<sup>28</sup>  $\gamma$  rays in <sup>17</sup>O. Although the Compton tails of coincident higher energy <sup>17</sup>O  $\gamma$  rays contribute to the background at 2.223 MeV, this cascade, for which the cross section is known, provides a very useful calibration of the gain, detection efficiency and energy resolution for the data accumulated over the 45 day measurement.

The area of the <sup>17</sup>O peak and the area of a possible peak at 2.223 MeV were obtained by summing over four channels (8 keV) at the correct energies and subtracting a background determined by averaging over 20 channels on either side of the peak. The area at 1.959 MeV is  $279 \pm 29$  counts and at 2.223 MeV the area is  $11 \pm 25$  counts. As discussed in Sec. II B, some cross-registration events are expected at 2.223 MeV even with the heavy metal shield in place and we calculated that  $19 \pm 4$  events are expected from this effect. Thus the net area from the <sup>1</sup>H( $n, \gamma\gamma$ )<sup>2</sup>H reaction is  $-8 \pm 25$  counts.

Because of the close target-detector geometry and the attenuation of the incident neutron flux by scattering in the target and absorption in the <sup>6</sup>Li, the absolute efficiency for coincidence detection is strongly dependent on the neutron beam profile and the target material. However, the <sup>1</sup>H( $n, \gamma\gamma$ )<sup>2</sup>H cross section limit can be determined by normalizing to the known <sup>16</sup>O( $n, \gamma$ )<sup>17</sup>O cross section if the relative  $\gamma$ -ray detection efficiencies are known. These relative efficiencies were determined from the measured intensities<sup>29-31</sup> of  $\gamma$  rays from neutron capture in graphite (carbon), Melamine (nitrogen), and lithium hydride (<sup>7</sup>Li) targets.

An independent measurement<sup>32</sup> was performed to confirm the only previously reported measurement<sup>28</sup> of the <sup>16</sup>O( $n, \gamma$ )<sup>17</sup>O cross section and branching ratios.

Branching ratios were measured for decay from the <sup>17</sup>O capturing state to the 871 keV level [(18  $\pm$ 3)%] and to the 3055 keV level [(82 $\pm$ 3)%]. The total cross section of <sup>16</sup>O( $n, \gamma$ )<sup>17</sup>O was determined to be 202 $\pm$ 27 µb. These results are in agreement with the results of Jurney and Motz<sup>28</sup> who measured branching ratios of 18% and 82% and a total cross section of 178 $\pm$ 25 µb. Averaging these measurements gives a cross section of 156 $\pm$ 16 µb for the 1088-2184-871 keV cascade in <sup>17</sup>O.

The net area of the sum peak at 2223 keV is related to the differential two-photon cross section through

$$\text{Yield} = k \int_{600 \text{ keV}}^{1620 \text{ keV}} dE_{\gamma} \int_{\Omega_{A}} d\Omega_{A} \int_{\Omega_{B}} d\Omega_{B} \frac{d\sigma_{2\gamma}(E_{\gamma}, \theta_{A}, \theta_{B})}{dE_{\gamma} d\Omega_{A} d\Omega_{B}} \left\{ \epsilon^{A}(E_{\gamma}) \epsilon^{B}(2223 - E_{\gamma}) + \epsilon^{B}(E_{\gamma}) \epsilon^{A}(2223 - E_{\gamma}) \right\},$$

where  $\epsilon^{A}(E_{\gamma})$  is the relative detection efficiency of detector A and k is the normalization constant.

The expression in brackets was found to vary by less than 10% for the energy range 600 keV  $\langle E_{\gamma} \rangle$  $\langle 1620 \text{ keV}$  and was assumed to be constant. For the *E*1-*E*1 cascade the predicted angular distribution is 1 + cos<sup>2</sup>( $\theta_A - \theta_B$ ); however, both detectors subtended large solid angles (see Fig. 1) and we estimate that an assumption of isotropic angular distributions would lead to an error of less than 10%. Consequently we assume an isotropic angular distribution and thereby avoid the problem of determining the directional efficiencies of the detectors. With these assumptions the above integral is proportional to the total two-photon cross section for the  $\gamma$  rays emitted in the energy range 600 keV  $\langle E_{\gamma} \rangle$  1620 keV.

The normalization constant k is determined from the yield of the sum peak from the 1088-871 keV cascade in <sup>17</sup>O. For this case an expression similar to that given above, but involving only the angular integration, can be used. The angular correlation for the <sup>17</sup>O cascade is isotropic since all the intermediate levels have spin  $j = \frac{1}{2}$ .

From the normalization constant k determined from the yield of the <sup>17</sup>O cascade, the measured relative efficiencies and the net intensity (-8±25) of the sum peak at 2.223 MeV we obtain

$$\sigma_{2\mathbf{v}} = -3 \pm 8 \ \mu \mathbf{b}$$

for 600 keV  $<\!E_{\gamma}\!<\!1620$  keV where the error  $\pm8~\mu{\rm b}$  corresponds to one standard deviation.

This corresponds to a branching ratio

$$\sigma_{2\gamma}/\sigma_{1\gamma} = (-0.8 \pm 2.5) \times 10^{-5}$$
.

Since a negative number for the cross section is unphysical we will adopt an upper limit of one standard deviation above zero. (8  $\mu$ b for  $\sigma_{2\gamma}$  and  $2.5 \times 10^{-5}$  for  $\sigma_{2\gamma}/\sigma_{1\gamma}$ .)

## **III. COMPARISON WITH OTHER EXPERIMENTS**

The present upper limit for the  ${}^{1}\text{H}(n, \gamma\gamma)^{2}\text{H}$  cross section at 85° ( $\sigma_{2\gamma} < 8 \ \mu \text{b}$ ) for 600 keV  $< E_{\gamma} < 1620$ 

keV is consistent with the early work of Arnold et al.<sup>12</sup> at 180° (<700 µb) for 1.2 MeV <  $E_{\gamma}$  < 1.6 MeV and the recent work of Wüst et al.<sup>24</sup> at 110° (-28 ±49 µb) for 46 keV <  $E_{\gamma}$  < 2177 keV. It is a factor of 44 lower than the value reported by Dress et al.<sup>13</sup> at 180° (350±50 µb) for 600 keV <  $E_{\gamma}$ < 1620 keV. This latter measurement was performed with two NaI detectors on opposite sides of the H<sub>2</sub>O sample. The present experiment was performed with Ge(Li) detectors subtending an angle of 85° at the target. We have already indicated that in the present measurement cross registration was reduced by shielding and the results of calculations<sup>26</sup> suggest it was an uncorrected problem in the earlier measurement.<sup>13</sup>

We also attempted to measure  $\sigma_{2\gamma}$  with the detectors at 180°. The sum spectrum so obtained is shown in the upper part of Fig. 5 and implies, if all events are assumed to be due to the  ${}^{1}\text{H}(n, \gamma\gamma){}^{2}\text{H}$  reaction, that  $\sigma_{2\gamma} = 200 \ \mu\text{b}$ . A second measurement with two Pb screens in front of the detectors (lower part of Fig. 5) reduced the yield in the sum peak by a factor of  $4.8 \pm 1.6$ .

The predicted attenuation for  $\gamma$  rays in the energy range 600 keV to 1620 keV is a factor of 3 if the events were due to the  ${}^{1}\text{H}(n, \gamma\gamma)^{2}\text{H}$  process and a factor of 5 if the events were due to cross registration caused by the scattering of the 2.223 MeV  $\gamma$  ray from one detector into the other. In addition,  $(81 \pm 29)\%$  of the events recorded in the sum peak at 2.223 MeV in Fig. 5 are predicted by calculations of cross registration<sup>26</sup> to be due to positron annihilation in flight.



FIG. 5. Sum spectra at 180° with and without Pb screens between the sample and detector, normalized to the same neutron fluence.

A strong angular correlation for the  ${}^{1}H(n, \gamma\gamma)^{2}H$ process might possibly explain the large ratio  $[\sigma_{2\gamma}(180^{\circ})/\sigma_{2\gamma}(85^{\circ}) \ge 25]$  of the cross sections at  $180^{\circ}$  and  $85^{\circ}$  measured with Ge(Li) detectors. However, in view of the large solid angle subtended by the detectors, this would be an extremely strong angular correlation. Because of the cross-registration calculations and the Pb screen measurements we reject this hypothesis and instead attribute the sum coincidence events seen at  $180^{\circ}$ in the present measurement and in the earlier measurement<sup>13</sup> to the annihilation in the detectors of positrons in flight.

## IV. COMPARISON WITH THEORY

The first of several recent theoretical papers on the subject of two-photon emission following thermal neutron capture was by Grechukhin<sup>14</sup> who obtained the estimate  $\sigma_{2\gamma} \approx 0.012 \ \mu$ b for the <sup>1</sup>H( $n, \gamma\gamma$ )-<sup>2</sup>H cross section. Following the apparently anomalous experimental results of Dress *et al.*,<sup>13</sup> there have been a number of calculations<sup>15-20</sup> which gave similar results although they use somewhat different approaches. However, in all calculations, the cross section is expected to be dominated by *E*1-*E*1 transitions from the <sup>3</sup>S<sub>1</sub> state and *M*1-*M*1 terms are found to be about 1000 times smaller.

Blomqvist and Ericson<sup>15</sup> used a gradient operator for the electric dipole interaction and obtained the result  $\sigma_{2\gamma} = 0.12 \ \mu b$ . (This value corresponds to 0.07  $\mu$ b for the energy range 600 keV <  $E_{\gamma}$  < 1620 keV.) Their approach is similar to that of Grechukhin<sup>14</sup> but they point out a missing factor of 8 in his calculation. Adler  $et \ al.^{20}$  employed a gradient operator and included nucleon-nucleon interactions to obtain the result  $\sigma_{2\gamma} = 0.096 \ \mu b$ . Hyuga and Gari<sup>16</sup> used a dipole operator with closure and obtained a value 0.083  $\mu$ b. Lee and Khanna<sup>18,19</sup> used a dipole operator to calculate  $\sigma_{2y}$ =0.12  $\mu$ b. They examined the various approaches and found that the calculation with the dipole operator is independent of the details of the wave functions, providing that the asymptotic behavior is correct and that the normalization of the deuteron wave function is consistent with low energy n-pscattering data. Blomqvist and Ericson<sup>15</sup> have recently refined their earlier calculation but again obtain the result  $\sigma_{2\gamma} = 0.12 \ \mu b$ .

The agreement among the various calculations is very good and gives confidence that any observed two-photon cross section larger than 0.12  $\mu$ b must arise from some process other than the *E*1-*E*1 two step transition via an intermediate <sup>3</sup>*P* state.

It has been suggested<sup>9</sup> that the contact gauge term  $[(e^2/2m)A(r_p)\cdot A(r_p)]$  could contribute to the

cross section. The matrix element for this process is proportional to the overlap between the deuteron and the  ${}^{3}S_{1}$  scattering wave functions which are normally thought to be orthogonal. However, in an attempt to explain a long standing discrepancy between the experimental and theoretical  $\sigma_{1y}$  Breit and Rustgi<sup>8</sup> suggested that this overlap might be nonzero due to the presence of mesons and isobars in a more complete description of the wave function. If this overlap were as large as the radial overlap between the deuteron and the  ${}^{1}S_{0}$ wave functions (i.e.,  $\langle D | {}^{3}S_{1} \rangle = \langle D | {}^{1}S_{0} \rangle$ ) then there would be a contribution of about 8% to the single photon cross section. However, more recent calculations<sup>4-7</sup> have indicated that most of the 8% discrepancy in  $\sigma_{1y}$  may be explained by exchange currents.

Prior to these recent calculations, Adler<sup>9</sup> calculated that an overlap of this size would result in a contribution (from the contact gauge term) to the two-photon total cross section for thermal neutrons of  $\approx 42 \ \mu$ b. Blomqvist and Ericson<sup>15</sup> recalculate  $\sigma_{2\gamma}$  from the contact gauge term to be 20  $\mu$ b for the assumption  $\langle D|^{3}S_{1}\rangle = \langle D|^{1}S_{0}\rangle$ . From the calculated<sup>15</sup> two-photon energy spectrum, 64% of the total  $\sigma_{2\gamma}$  (or 13  $\mu$ b) falls in our experimental energy range, 600 keV  $\leq E_{\gamma} \leq 1620$  keV. Our observed upper limit of 8  $\mu$ b is lower than the  $\sigma_{2\gamma}$ calculated using this nonorthogonality assumption.

In addition, Blomqvist and Ericson present dimensional arguments which suggest that  $\langle D | {}^{3}S_{1} \rangle \leq \frac{1}{30} \langle D | {}^{1}S_{0} \rangle$  thus setting an upper limit to the contribution to  $\sigma_{2\gamma}$  from the contact gauge term of about 0.03  $\mu$ b. Moreover, other authors<sup>10,11</sup> have stated that the suggestion of nonorthogonality be-tween the  ${}^{3}S_{1}$  and deuteron wave functions is untenable.

Other possible contributions to  $\sigma_{2\gamma}$  have been calculated by various authors and found to be negligible compared to 0.1  $\mu$ b. Bernabéu and Tarrach<sup>17</sup> consider bremsstrahlung accompanying singlephoton emission and find  $\sigma_{2\gamma} \approx 1.5 \times 10^{-4} \ \mu$ b. Friar<sup>21</sup> has shown that pion exchange terms do not contribute significantly to  $\sigma_{2\gamma}$ . Blomqvist and Ericson<sup>15</sup> estimate the cross section for two-photon emission by a virtual  $\pi^{0}$  to be  $\approx 3 \times 10^{-9} \ \mu$ b.

In conclusion it can be stated that the present experimental limit on  $\sigma_{2\gamma}$  is in agreement with conventional theories and has reached the level where restrictions are being imposed on possible contributions to  $\sigma_{2\gamma}$  and  $\sigma_{1\gamma}$  from more speculative effects such as a possible nonorthogonality of the  ${}^{3}S_{1}$  scattering state and the deuteron state wave functions. Although we are still far from the predicted cross section of 0.12  $\mu$ b, each improvement in sensitivity will increase our confidence in current theories of the np electromagnetic interaction.

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