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Further Evidence for the Nonexistence of Particle-Stable Tetraneutrons

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A search was made for the occurrence of particle-stable tetraneutrons in the fast-deuteron-induced fission of uranium. This process is known to give a high yield of alphas and tritons. In order to deduce the presence of tetraneutrons, the following hypothetical reactions were investigated: $N^{14}(n^4,n)N^{17}$, $O^{16}(n^4,t)N^{17}$, $Mg^{26}(n^4,2n)Mg^{28}$, $Rh^{103}(n^4,2n)Rh^{105}$, $Bi^{209}(n^4,n)Bi^{212}$ and $Bi^{209}(n^4,2n)Bi^{211}$. No evidence for tetraneutrons was found. The upper limits of tetraneutron yields per alpha obtained from the above reactions are: 2×10^{-8}, 3×10^{-4}, 3×10^{-5}, 3×10^{-4}, 1×10^{-6}, and 1×10^{-8}, respectively. It seems reasonable to conclude from these results that the existence of tetraneutrons is most unlikely.

As a consequence of experimental results from the $He^4(\gamma,\pi^+)\rightarrow t+n$ reaction, it has been suggested that there is a low-lying resonant state in the n-t system at about 4 MeV above binding.[1] Since this state could not be observed in n-t scattering,[2] it has been interpreted as a state with isotopic spin[3] $T=2$. On the basis of this conclusion one would expect the existence of a particle-stable system of four neutrons bound by about[3] 4.5 MeV. However, reinterpretation of the experimental results shows that it is difficult to deduce from the hitherto existing data whether or not there is an H^4 state present in the reaction products.[4,5] In a recent experiment, an upper limit of 15% was obtained for the production of an H^4 final state.[6] The possible occurrence of He^8 and pairing energy arguments cast some doubt upon the stability of the tetraneutron, although the suggestion in favor of it cannot be rejected entirely.[7] Symmetry considerations allow the conclusion that the proposed $T=2$ resonance state implies the $T=1$ state of H^4 to be bound.[8] However, no H^4 was found in several searches.[9]

The problem of the states n^4 and H^4 is closely connected with the problem of the excited states of the He^4 nucleus and the existence of[7,10] H^5. A He^4 level at about[11,12] 20.1 MeV with[13] $T=0$ seems to be well established. In a recent paper a second excited state has been proposed at about[12] 21.2 MeV. It can be either a $T=0$ or a $T=1$ state. On account of isotopic spin conservation, all experiments up till now concerning the He^4 level structure cannot provide information on

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states with $T=2$. With one exception[14] searches for particle-stable H^5 were unsuccessful.[15]

In this note a search for the occurrence of tetraneutrons in the fast-deuteron-induced fission of uranium is described. The measurements may be of use in clarifying the experimental situation for the four-nucleon system. A search for tetraneutrons in the thermal-fission process had a negative result.[8] If tetraneutrons exist at all, the yield in the fast deuteron-induced fission is expected to be about two orders of magnitude higher than in thermal fission. This assumption is reasonable because of the much higher yield of alphas and tritons.[16]

A natural-uranium target was bombarded with 4 μA of 50-MeV deuterons in the Karlsruhe isochronous cyclotron. In order to deduce the presence of n^4 the following hypothetical reactions were investigated: $N^{14}(n^4,n)N^{17}$, $O^{16}(n^4,t)N^{17}$, $Mg^{26}(n^4,2n)Mg^{28}$, $Rh^{103}(n^4,2n)Rh^{105}$, $Bi^{209}(n^4,n)Bi^{212}$ and $Bi^{209}(n^4,2n)Bi^{211}$. In none of these experiments was any evidence found for the existence of particle-stable tetraneutrons. The only information we are able to deduce from the experimental data is an upper limit of the number of n^4 produced per fission. In Table I the results are summarized together with the assumed n^4 cross sections. Taking into account the influence of binding energy the cross section values seem to be reasonable from (α,n), (α,p), (α,t), and $(\alpha,2n)$ cross sections in the mass region of the target nuclei.

TABLE I. Upper limits of tetraneutron yields.

No.	Reaction	Assumed $\sigma_{n^4,x}$ (mb)	n^4 yield per fission	n^4 yield per alpha	n^4 yield per triton
1	$N^{14}(n^4,n)$	50	$<5\cdot10^{-9}$	$<2\cdot10^{-8}$	$<5\cdot10^{-7}$
2	$O^{16}(n^4,t)$	40	$<1\cdot10^{-4}$	$<3\cdot10^{-4}$	$<1\cdot10^{-2}$
3	$Mg^{26}(n^4,2n)$	100	$<1\cdot10^{-5}$	$<3\cdot10^{-5}$	$<1\cdot10^{-3}$
4	$Rh^{103}(n^4,2n)$	500	$<1\cdot10^{-4}$	$<3\cdot10^{-4}$	$<1\cdot10^{-2}$
5	$Bi^{209}(n^4,n)$	50	$<4\cdot10^{-7}$	$<1\cdot10^{-6}$	$<4\cdot10^{-5}$
6	$Bi^{209}(n^4,2n)$	500	$<3\cdot10^{-9}$	$<1\cdot10^{-8}$	$<3\cdot10^{-7}$
1[a]	$N^{14}(n^4,n)$	50	$<2\cdot10^{-8}$	$<4\cdot10^{-6}$	$<2\cdot10^{-4}$
2[a]	$Al^{27}(n^4,t)$	40	$<5\cdot10^{-9}$	$<1\cdot10^{-6}$	$<5\cdot10^{-5}$

[a] Observed for thermal fission (Ref. 8).

In the first experiment, nitrogen samples were irradiated in the form of tetrazole N_4CH_2 and ammonium azide NH_4N_3. The occurrence of the $N^{14}(n^4,n)N^{17}$ reaction could be examined by looking for the 4.1-sec delayed-neutron activity of N^{17}. This technique provides excellent discrimination against other reaction products. In a pneumatically operated rabbit system, 1-g samples in polyethylene containers were irradiated for 20 sec at a point 3.3 cm from the uranium target. The samples were counted in a distant low-background assembly at 2.6-sec intervals. The neutron detector consisted of 17 $B^{10}F_3$ counters in a paraffin pile having an over-all efficiency of about 7%.

For the $O^{16}(n^4,t)N^{17}$ reaction, the same technique was applied. In order to determine the interference from the (n,p) and (n,d) reactions on the rare isotopes O^{17} and O^{18}, two "rabbits" containing D_2O of different oxygen isotopic composition were irradiated alternately. The accuracy of this experiment was limited by the uncertainties in the average (n,p) and (n,d) cross sections.

In the third experiment, a 5-g sample of MgO was irradiated for 5 h at a point 4.5 cm from the uranium target. A radiochemical separation of Mg was then performed to eliminate the high Na^{24} activity produced by the (n,p) process on Mg^{24}. The occurrence of the $Mg^{26}(n^4,2n)Mg^{28}$ reaction was examined by looking for the 1.35- and 1.78-MeV γ transitions following the β decay of 21.3-h Mg^{28} and 2.3-min Al^{28}, respectively. The sample was counted for 8 h with a 4-in.×5-in. NaI(Tl) scintillation detector.

For studying the $Rh^{103}(n^4,2n)Rh^{105}$ reaction a Rh foil, 60 μ thick and 1.6 cm in diameter, was irradiated for 5 h at a position 3 cm from the cyclotron target. The beta- and gamma-ray spectra of the sample were followed for a period of several days in a beta proportional counter and a 3-in.×3-in. NaI(Tl) detector, respectively. In the beta measurement, interference was observed from the 5% β^+ activity of 21-h Rh^{100} produced by the $(n,4n)$ reaction. The accuracy of the γ-ray spectrum analysis was limited by the presence of 4.5-day Rh^{101} which results from the $(n,3n)$ process and which has a gamma line very close to the 319 keV transition following the β decay of 35-h Rh^{105}.

The occurrence of the Bi^{209} reactions was examined by looking for the α activity of the product nuclei 2.15-min Bi^{211} and 60.5-min Bi^{212}. The target nuclide Bi^{209} has the advantage that short-time neutron irradiation cannot induce measurable α activities. Samples were prepared by evaporating layers of Bi about 100 μ thick on thin 4×4-cm copper foils. After 10 min of irradiation at a point 3.3 cm from the uranium target, these foils were counted for 8 min with a thin 2-in.-diam ZnS(Ag) scintillation screen.

Considering the absence of a Coulomb barrier for the tetraneutron, this particle should occur with a frequency comparable with that of alphas and tritons in spite of the much lower binding energy.[8] Therefore, it seems reasonable to conclude from Table I that the existence of tetraneutrons is most unlikely. As a consequence, the observed resonance state[1,3] in H^4, if it exists at all, most probably is not a $T=2$ state. Furthermore, the first He^4 state with $T=2$ should have an energy >29 MeV.

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Search for Tetraneutrons Using a Recoil Proton Detector

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In a recent paper[1)] we described a search for the occurrence of particle-stable tetraneutrons in the fast deuteron-induced fission of uranium. In these experiments several hypothetical reactions of the types (n^4,n), $(n^4,2n)$ and (n^4,t) were investigated for deducing the presence of tetraneutrons. The experimental data suggest the conclusion that the existence of these particles is most unlikely. In order to attach additional weight to our results it was deemed useful to perform a further experiment employing a different technique. In the present report a search for n^4 is described using a recoil proton detector and the time-of-flight method. Again, the fast deuteron-induced fission of uranium was used as a possible source of tetraneutrons. This process is known to give a high yield of alphas and tritons. Considering the absence of a Coulomb barrier for the tetraneutron this particle should occur with a comparable frequency, in spite of the much lower binding energy. A summary of the experimental and theoretical background for the four-nucleon system and of the corresponding literature is given in our previous paper.

Assuming that tetraneutrons undergo elastic collisions in a hydrogenous medium it follows from collision kinematics that the maximum energy transfers for tetraneutrons and neutrons of equal velocity are correlated by $E(n^4) = 2.56\ E(n)$. Thus, in recoil spectra measurements with selection of a fixed time-of-flight two superimposed spectra with well separated maximum energies are expected provided that tetraneutrons are produced in the source.

The block diagram of the experimental setup used in the present measurements is shown in fig. 1. A natural uranium target was bombarded with 100 nA of 50 MeV deuterons from the external beam of the Karlsruhe isochronous cyclotron. The deuteron pulses had a repetition rate of 33 Mc. Therefore, the detector had to be placed close to the target. A distance of 25 cm was used. The detector was a 5 mm Ø x 0.5 mm NE 213 liquid scintillator mounted on a 56 AVP photomultiplier. Pulse-shape discrimination was employed in order to eliminate pulses arising from time-uncorrelated gamma radiation. The circuit is based on the space charge limitation method proposed by Owen[2, 3]. A fast output signal started a tunnel diode time-to-pulse height converter. By a differential discriminator a flight time corresponding to 1.2 MeV neutron energy was selected. The overall energy resolution of the time-of-flight circuit was better than 20 %. Because of the large time jitter of the discriminators the coincidence resolving time was set to 2 μsec. Spectra were analysed using a 400 channel pulse-height analyzer.

A recoil spectrum corrected for time-uncorrelated background is shown in fig. 2 No evidence was found for the occurrence of a superimposed spectrum with higher maximum energy. The results of our previous experiments are thus confirmed. The only information we are able to deduce from the data is an upper limit for the numbers of tetraneutrons produced per fission. This limit was calculated to be 3×10^{-3}. The upper limits for the numbers of tetraneutrons per alpha and triton are 9×10^{-3} and 3×10^{-1}, respectively.

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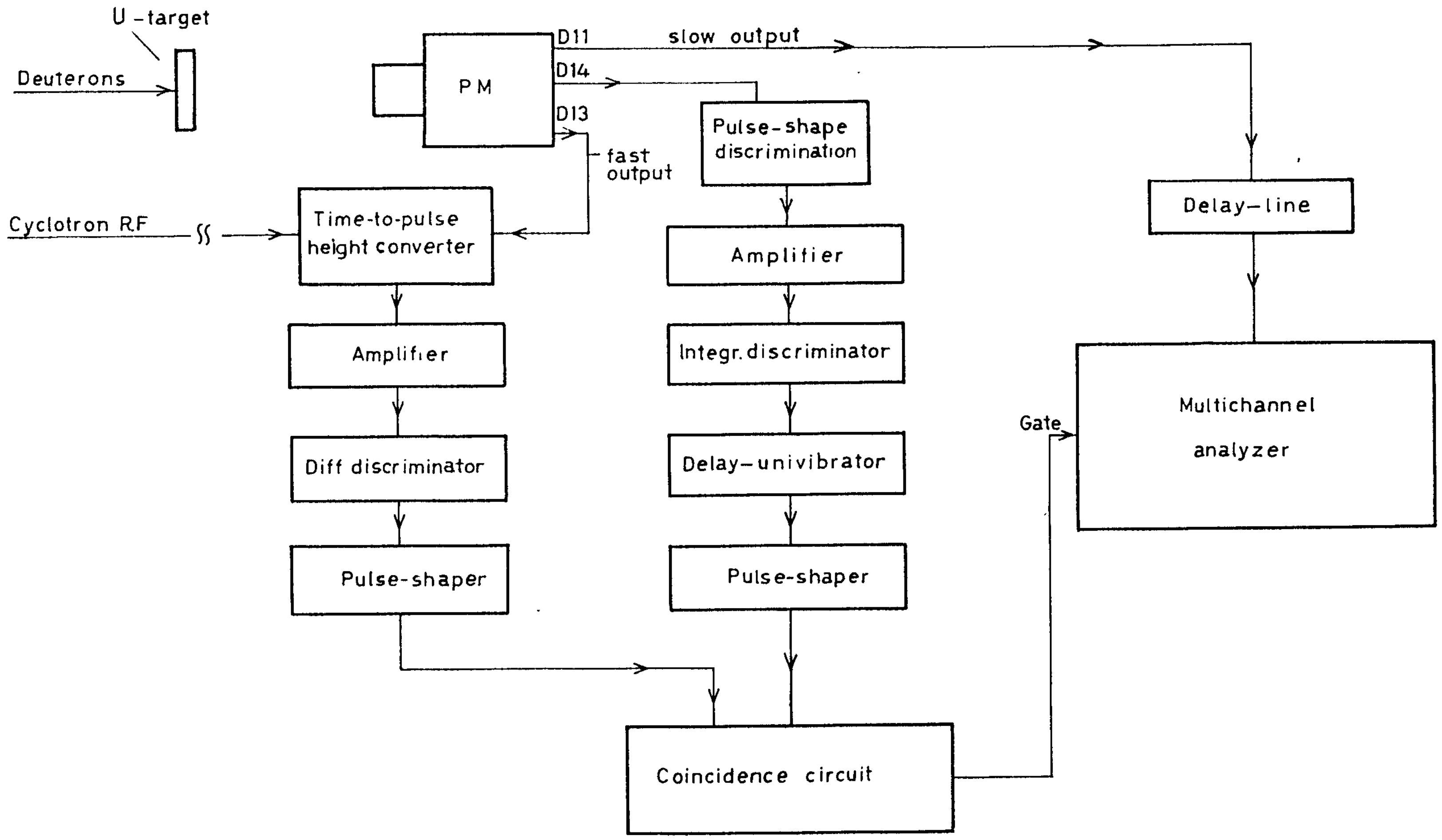

Fig.1 Block diagram of the experimental setup

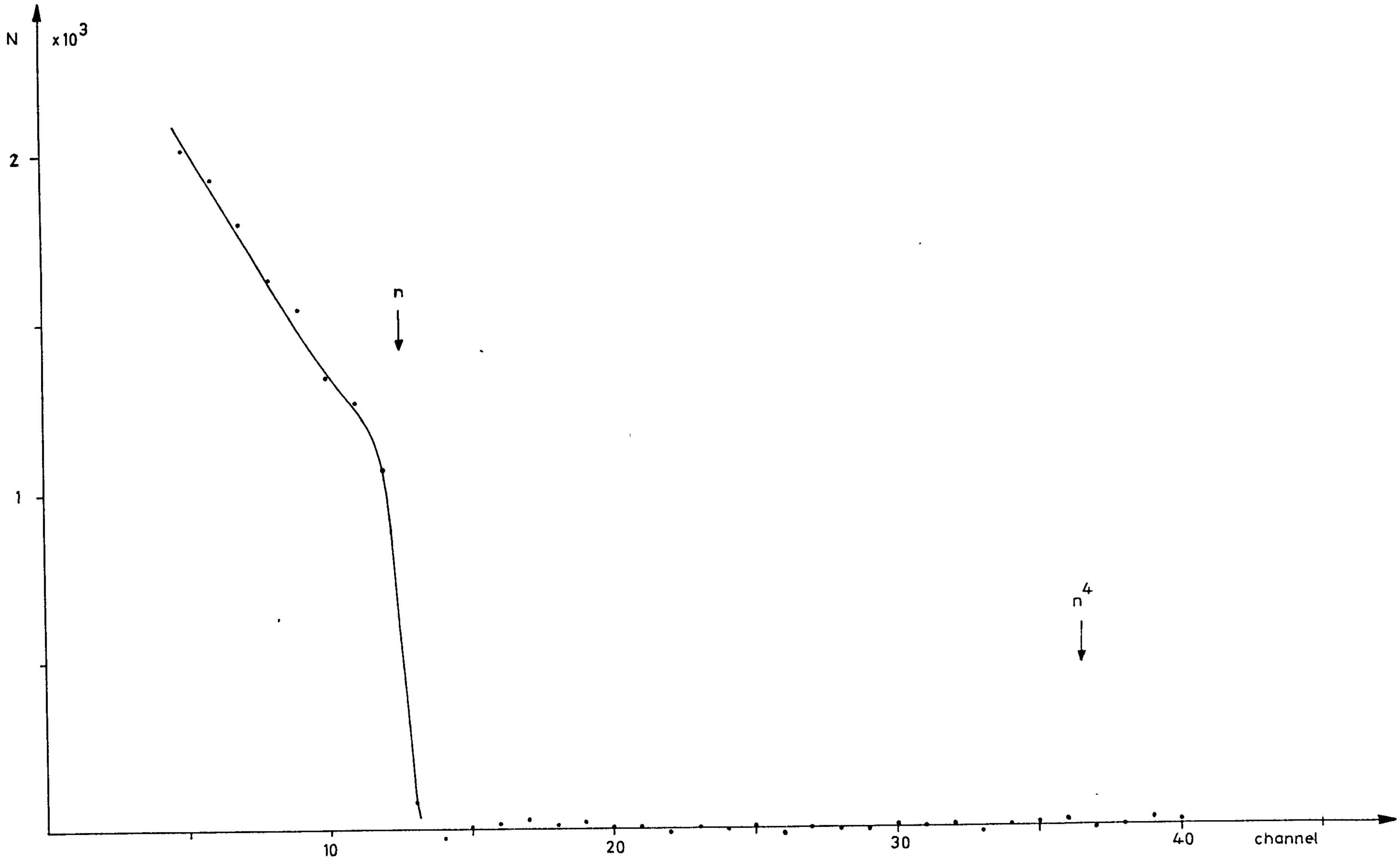

Fig. 2 Proton recoil spectrum corresponding to a neutron energy of 1.2 MeV