of these direct interaction processes would be of considerable interest, since one might hope to learn more of the details of the particle substructure of nuclei from them.

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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: $2\times10^{-8}$, $3\times10^{-4}$, $3\times10^{-5}$, $3\times10^{-4}$, $1\times10^{-6}$, and $1\times10^{-8}$, 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 $\mu$A 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 $(\alpha,n)$, $(\alpha,p)$, $(\alpha,t)$, 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 $\gamma$ transitions following the $\beta$ 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 $\mu$ 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% $\beta^+$ activity of 21-h $Rh^{100}$ produced by the $(n,4n)$ reaction. The accuracy of the $\gamma$-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 $\beta$ decay of 35-h $Rh^{105}$.

The occurrence of the $Bi^{209}$ reactions was examined by looking for the $\alpha$ 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 $\alpha$ activities. Samples were prepared by evaporating layers of Bi about 100 $\mu$ 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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