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BETA-DELAYED THREE-NEUTRON RADIOACTIVITY OF ^{11}Li

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ABSTRACT

A new radioactive decay mode, beta-delayed three neutron emission, has been observed in ^{11}Li in an intensity $p_{3n} = 1.8 \pm 0.2\%$. The single and double neutron branches are $p_{1n} = 82 \pm 7\%$ and $p_{2n} = 3.9 \pm 0.5\%$.

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Beta-delayed emission of two neutrons has recently been observed in the decay of ${}^7\text{Li}$ [1] and of the sodium isotopes ${}^{30-32}\text{Na}$ [2]. In the present paper is reported the extension of the neutron time-correlation techniques used in the previous works to the detection of beta-delayed three neutron emission.

The isotope ${}^7\text{Li}$ with a half-life of 8.6 ± 0.2 ms is ideally suited for a search for 3n emission. It has a Q_β value of 20.7 MeV [3] and the thresholds for break up into three neutrons lie at 8.888 MeV (residual two ${}^4\text{He}$) and at 8.979 MeV (residual ${}^9\text{Be}$) [4]. The decay by 2n emission seems to proceed largely through the broad ${}^7\text{Be}$ resonance at 8.84 MeV [5], as is indicated by the shape of the spectrum of beta-delayed neutrons [1]. The first excited state of ${}^9\text{Be}$ (1.68 MeV, $\frac{1}{2}^+$) corresponding to an excitation energy of 8.995 in ${}^7\text{Be}$ is broad and decays by neutron emission. Thus, 3n emission could proceed not only through high-lying states in ${}^7\text{Be}$, but also through tails of resonances in ${}^9\text{Be}$ and ${}^7\text{Be}$ near the threshold for three-neutron emission.

The radioactivity was produced by bombarding a target of uranium carbide at 2000°C with a 0.2-2 μA proton beam of 600 MeV from the CERN synchro-cyclotron. In order to minimize the effect of the macro-structure of the beam, the cyclotron RF was operated in the 1 : 1 mode corresponding to a 3 ms spacing between beam pulses. The ${}^7\text{Li}$ radioactivity was separated on-line in the ISOLDE facility and directed as an ion beam to the centre of a paraffin-filled 4π neutron counter.

The neutron counter [1,2] now was equipped with a total of 12 ${}^3\text{He}$ tubes, which increased the efficiency to $20 \pm 1\%$ as determined with a 47.7 g sample of ${}^{238}\text{U}$. (Note, however, that the neutron detection efficiency is expected to depend appreciably on the neutron energy, cf. e.g. ref. [6].) The residence time in the detector, as determined from beta-neutron coincidences on ${}^9\text{Li}$ was exponentially distributed with a mean of 89 ± 1 μs . The neutron counters were connected in parallel and fed into a micro-processor unit that allowed the arrival times of individual neutrons to be read by a "flying clock" with a precision of 1 μs . The correlation analysis could therefore be performed in a playback mode from a magnetic tape.

The rate of triple events and the histograms (Fig. 1) taken on-line with strongly reduced proton beam intensity clearly indicated the presence of true triple events. The presence of random triple correlations due to the combination of single and double events, however, necessitated a more detailed analysis, of which we outline the main features.

Let a source of (constant) beta disintegration rate D decay through channels involving the emission of i neutrons with the probabilities p_{in} per beta decay. In most cases, however, a single neutron only will be detected, and it is therefore

convenient to define the rates R_s of actual correlated events of s counts in the data string. If each multiplicity is characterized by an energy-independent average efficiency ϵ_i , we may write

$$R_s = D \sum_{i=s}^{\infty} p_{in} \binom{i}{s} \epsilon_i^s (1-\epsilon_i)^{i-s} \quad (1)$$

where $\binom{i}{s}$ is the binomial coefficient. It is clear that the R_s are the quantities directly determined from a correlation analysis of the data and that the ratios of the p_{in} 's may subsequently be found by solving the equations (1).

We have chosen, as a strategy for detecting correlations, to examine a time interval θ following an initial event. If $(q-1)$ neutrons have been recorded during this time, we register the event as a q -fold event. For an individual count belonging to the event, the detection probability per unit time is $\lambda e^{-\lambda t}$, where λ^{-1} ($= 89 \mu s$) corresponds to the mean residence time in the counter. With this strategy the (true) counting rate of q -fold events is

$$M_q^{(t)} = \sum_{s=q}^{\infty} R_s r_{s,q} \quad (2)$$

where $r_{s,q}$ is the probability of exactly $q-1$ counts out of $s-1$ possible will fall inside the time window

$$r_{s,q} = \binom{s-1}{q-1} (1-e^{-\lambda\theta})^{q-1} e^{-\lambda\theta(s-q)} \quad (3)$$

(It may furthermore be necessary to take into account that the undetected counts will re-appear as a second event with multiplicity $s-q$.)

The random events $M_q^{(r)}$ can be calculated ^{*)} as combinations of events of lower multiplicity. It is now clear that it is possible from the measured $M_q = M_q^{(t)} + M_q^{(r)}$ + small corrections to solve the equations for the R_s .

*) For completeness, we give here the expressions for the rates of random doubles

$$M_2^{(r)} = R_1^2 \theta e^{-R_1\theta}$$

and triples

$$M_3^{(r)} = R_1 R_2 \theta e^{-R_1\theta} \left[2e^{-\lambda\theta} - \frac{1-e^{-\lambda\theta}}{\lambda\theta} \right] + \frac{1}{2} R_1^3 \theta^2 e^{-R_1\theta}$$

where the terms, of course, also must appear as negative corrections to the rates of events with lower multiplicities. (The three terms correspond to (1^2) , $(2-1)$ and (1^3) random coincidences.)

Fluctuations in the data rate can be taken into account through replacements of the type

$$\langle R_1 R_2 \rangle = \langle R_1 \rangle \langle R_2 \rangle \frac{\langle D^2 \rangle}{\langle D \rangle^2} \quad (4)$$

and similar for R_1^2 and R_1^3 where the symbol $\langle \rangle$ denotes time average. The rate fluctuations due to the 3 ms spacing between the pulses of the proton beam are damped by the delay in the target so that this effect can be neglected. Long-term variations in the data rate were corrected for by computing D , D^2 and D^3 for each buffer of ~ 1350 counts and by averaging over the whole run. The biggest correction was for the run at 7.9 c/s (see below), for which we found $\langle D^2 \rangle / \langle D \rangle^2 = 1.011$ and $\langle D^3 \rangle / \langle D \rangle^3 = 1.032$.

The raw experimental data were first corrected for the counter dead time of 5 μ s. At a correlation time $\theta = 228 \mu$ s this correction amounted to 5.9% for the doubles and 17.2% for the triples. The equations for R_s were subsequently solved including terms up to $s = q = 3$ only, which is sufficient here. The calculated R_s subsequently, were converted to ratios of the p_{in} 's, by means of (1). It was assumed *) that the ϵ_1 could be replaced by the single value $\epsilon = 0.20$.

As the evaluation is very sensitive to the precision with which the correction for randoms can be carried out, the analysis was checked in several ways.

- (i) Random data taken with a γ -source and with ^9Li neutrons were analyzed and it was found that the corrections for random events were accurate to typically 3-7%.
- (ii) The neutron count rate in the ^{11}Li experiments was varied from 7.9 c/s to 202.8 c/s without any appreciable change in the results (Table 1). This is a very sensitive test as the random events vary with the square or the cube of the rate.
- (iii) The analysis time θ was varied between 79.8 and 456 μ s, again without any detectable effect (Table 1).
- (iv) The time distributions shown in Fig. 1 were calculated from the results of the data analysis and evidently agree exactly with experiment.

The results given in Table 1 could change markedly if the different multiplicities turn out to have very different energy spectra so that the ϵ_1 (eq. 1) become very different. We believe that other sources of systematic errors are excluded beyond the level of the individual errors in Table 1. For this latter reason a weighted average would not be meaningful, and we propose as adopted values

*) The approximation of constant efficiency is clearly the best that can be done in the absence of measured neutron coincidence spectra. Note that a recalculation with new ϵ_1 could be done from (1) alone.

$p_{2n}/p_{1n} = (4.8 \pm 0.5) \cdot 10^{-2}$ and $p_{3n}/p_{1n} = (2.2 \pm 0.2) \cdot 10^{-2}$. The p_{in} values may be put on an absolute scale by means of the total probability for neutron emission $p_n = \sum_{i=1}^{\infty} i p_{in}$. For ^{11}Li this quantity has been determined by Roeckl et al. [7] and more recently by others [8,9], and it now seems clear that all experiments consistently point to a value near 100%. We use here the value $p_n = 95 \pm 8\%$ [8]. From this one finds for ^{11}Li $p_{1n} = 82 \pm 7\%$, $p_{2n} = 3.9 \pm 0.5\%$ and $p_{3n} = 1.8 \pm 0.2\%$. In hindsight, these results are in agreement with the previous two-neutron experiment [1], which actually determined the quantity $p_{2n} + 2.8 p_{3n} = 0.15 \pm 0.05$ now expressed in terms of the new p_n value for ^{11}Li .

The experiment represents the first observation of a new radioactive decay mode : beta-delayed three-neutron emission. It is furthermore noteworthy that the residual nucleus itself is unstable so that the process detected in the present work effectively is a break up into five nuclear particles ($2\alpha + 3n$).

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Table 1

Relative two- and three-neutron branching ratios

Singles rate c/s	$10^2 \times p_{2n}/p_{1n}$	$10^2 \times p_{3n}/p_{1n}$
7.9 ^{a)}	5.2 ± 0.6	2.2 ± 0.2
22.9 ^{a)}	4.5 ± 0.3	2.45 ± 0.09
45.5 ^{a)}	4.6 ± 0.5	2.35 ± 0.2
202.8 ^{b)}	5.0 ± 1.2	2.0 ± 0.4
Correlation time θ μs ^{c)}		
79.8	5.2 ± 1.0	2.2 ± 0.3
114	5.9 ± 0.6	2.0 ± 0.2
228	5.2 ± 0.6	2.2 ± 0.2
456	5.0 ± 0.6	2.39 ± 0.16

a) $\theta = 228 \mu s$

b) $\theta = 114 \mu s$

c) Singles rate $R = 7.9$ c/s. The results shown correspond to the analysis of the same set of data for four different values of the correlation time θ .

FIGURE CAPTION

Fig. 1 : Distribution of the time interval between the first and the second neutrons for events registered as doubles and triples with a correlation time $\theta = 228 \mu\text{s}$. The data are from a 12 h run with an average neutron count rate of 22.9 c/s. The theoretical curves showing the total number of events and the contribution from random events (r) do not represent a fit : they have been calculated on an absolute scale from the results of the analysis described in the text. Note that the triples events clearly show the $e^{-2\lambda t}$ dependence expected theoretically.

Fig. 1