

BETA-DELAYED TWO-NEUTRON DECAY STUDIES FOR $^{96-99}\text{Rb}^*$

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Abstract

Beta-delayed two-neutron emission from $^{96-99}\text{Rb}$ has been studied by use of the neutron-neutron time correlation technique. Time interval distributions for neutron pulses from a polyethylene moderated neutron counter were measured for mass-separated sources. Coincident neutrons were identified in the time interval distribution by a component having the characteristic residence time of 35 μs for this counter. At mass 98 a coincidence rate well above the correlated background results in a P_{2n} for ^{98}Rb of $(0.060 \pm 0.009)\%$. Upper limits (2σ) for P_{2n} of 0.004, 0.008, and 0.024% are reported for ^{96}Rb , ^{97}Rb , and ^{99}Rb , respectively.

1. Introduction

Calculations based on semiempirical mass formulas predict that beta-delayed two-neutron emission should be energetically possible for many nuclides near the presently known limits of particle stability. From the mass formulas suggested by Monahan and Serduke,¹⁾ one predicts²⁾ that beta-delayed two-neutron emission is marginally allowed for ^{96}Rb , forbidden for ^{97}Rb , and allowed for ^{98}Rb and ^{99}Rb . The relevant beta decay energies (Q_β), two-neutron binding energies in the daughter nuclide (B_{2n}), and energy windows ($Q_\beta - B_{2n}$) are given in Table I.

TABLE I. Calculated Values of Q_β , B_{2n} , and Energy Window for Beta-Delayed Two-Neutron Emission^a

Nuclide	Q_β (MeV)	B_{2n} (MeV)	$Q_\beta - B_{2n}$ (MeV)
^{96}Rb	11.0	10.8	0.2
^{97}Rb	9.1	10.5	-1.4
^{98}Rb	12.2	10.3	1.9
^{99}Rb	10.1	9.6	0.5

a Based on mass formulas in Reference 1.

To discover whether beta-delayed two-neutron emission does occur in the neutron-rich Rb isotopes, we used the TRISTAN on-line isotope separator at the High Flux Beam Reactor at Brookhaven National Laboratory.³⁾ For these experiments, the TRISTAN facility had a surface ionization source containing enriched ^{235}U located in a beam of thermal neutrons. With a Re surface at high temperatures ($>1800^\circ\text{C}$), the ion beams at masses 96 to 99 are mixtures of Rb and

Sr fission products. The ion beams were deposited at the center of a high efficiency neutron counter (SNC - SOLAR Neutron Counter) described previously.^{4,5)}

The neutron-neutron time correlation technique described by Azuma, et al.^{6,7)} and Detraz, et al.⁸⁾ was adopted for the present work. The time interval distribution for neutron pulses was measured for each of the Rb isotopes. From the time interval distribution one obtains the ratio of single to double neutron emission probabilities (P_{1n}/P_{2n}). One then uses the literature values of P_n to obtain P_{2n} .

To derive an expression for P_{1n}/P_{2n} , we first define C as the total neutron count rate

$$C = D E P_{1n} + 2 \cdot D E P_{2n} \quad (1)$$

where D = beta disintegration rate
E = neutron detection efficiency.
We then define I_S as the coincidence counting rate as determined by the rate for short time intervals in the time interval distribution.

$$I_S = D E^2 P_{2n} \quad (2)$$

In Equations 1 and 2 we have assumed that the efficiency for detecting uncorrelated neutrons is the same as that for coincident neutrons. We then divide Equation 1 by Equation 2, rearrange, and obtain P_{1n}/P_{2n} .

$$\frac{P_{1n}}{P_{2n}} = E \left(\frac{C}{I_S} - \frac{2}{E} \right) \quad (3)$$

The experimental determination of C/I_S is discussed below.

2. Experimental

The neutron counter (SNC) consists of 40 tubes filled with 4 atm of ^3He arranged in three concentric rings in a polyethylene moderator. The efficiency of each ring as a function of neutron energy has been measured with calibrated photoneutron sources. For a spectrum of neutrons with an average energy of 500 keV, the total efficiency is $(59 \pm 5)\%$. The total efficiency decreases slowly over the range of 100 to 1000 keV but stays within the uncertainty limits quoted for the absolute efficiency at 500 keV. The average energy of the two-neutron emission is unknown, but is expected to lie with this range.

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Pulses from the SNC were sent to a specially designed NIM module which replaced the ADC in the TRISTAN data acquisition system. The time interval between successive pulses was converted to an address which was then used to increment the distribution of time intervals. The NIM module contained a 40 word buffer memory which derandomized the memory transfer operation. All time intervals which exceeded the allowed range were stored in the last channel of memory (8192) so all time intervals were counted.

The time interval distribution $N(t)$ for a random source having a constant count rate λ is derived from Poisson's distribution which gives the expression

$$N(t) = \lambda e^{-\lambda t} \quad (4)$$

The mean time between pulses is $1/\lambda$. If the random event rate is low (<200 counts/sec), the time interval distribution is almost flat on a time scale of <0.5 ms. If coincident neutrons are generated within the SNC, the time interval distribution will contain a second exponential component with a mean time given by the residence time of neutrons in the counter.

The residence time was measured by two different techniques. The first technique involves the measurement of the time interval distribution from a spontaneous fission source - either a large quantity of natural ^{238}U located at the source position inside the SNC or a ^{252}Cf source located outside the SNC. This technique suffers from the fact that spontaneous fission neutrons have a multiplicity distribution extending beyond two neutrons per event. Thus the time interval distribution has even shorter exponential components corresponding to higher multiplicity events.⁶⁾ The time interval distribution of ^{238}U is shown in Figure 1. Although a single exponential plus background gives a reasonable fit to the data, there is evidence for a small deviation at short time intervals.

The second technique for residence time measurements is to record the time intervals between beta particles and neutrons from a beta-delayed neutron source. A standard time-to-amplitude converter (TAC) was triggered by start pulses from a beta detector inside the SNC and by stop pulses from the SNC. The TAC output was calibrated by introducing known delay times from a variable gate-and-delay generator into the stop input of the TAC. The time interval distribution for a beta-start neutron-stop TAC experiment with a ^{96}Rb source is shown in Figure 2. The solid lines show the result of a least squares fit using two exponential components. The residence times measured by the different techniques are consistent with each other as shown in Table II. The relatively short residence time of $35 \mu\text{s}$ is a consequence of the large number of ^3He tubes in the counter volume and is advantageous for distinguishing coincident neutrons from the random background.

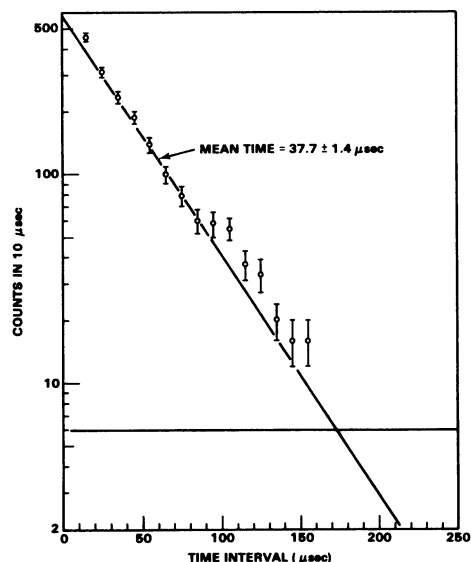


FIGURE 1. Time Interval Distribution for ^{238}U Source Located in Center of SNC. Lines indicate best fit of two exponential components.

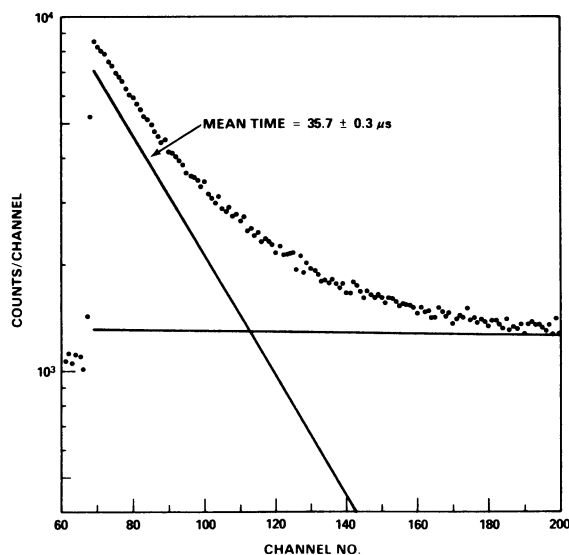


FIGURE 2. Residence Time Measurement by Beta-Start Neutron-Stop TAC Technique with ^{96}Rb Source. Fixed delay of $100 \mu\text{s}$ for neutron-stop signal put zero time in Channel 69. Calibration = $1.39 \mu\text{s}/\text{ch}$. Lines indicate best fit of two exponential components.

TABLE II. Residence Time of Neutrons in SNC

Source	Location	Method	Total Count Rate (n/s)	Residence Time (μ s)
^{238}U	Internal	Time Interval Module	6.3	37.7 ± 1.4
^{252}Cf	External	Time Interval Module	865.0	35.2 ± 2.3
^{96}Rb	Internal	β -n TAC	81.6	35.7 ± 0.3
^{97}Rb	Internal	β -n TAC	5.5	33.0 ± 1.0
^{98}Rb	Internal	β -n TAC	10.6	34.6 ± 1.8
			Adopted Value	35.0 ± 2.0

The total neutron count rate, C, was obtained as the integral of the time interval distribution divided by the length of the run. Note that the number of time intervals is equal to the number of neutron counts minus one. This count rate was corrected by subtracting the background count rate and multiplying by the fraction of the neutrons due to the Rb precursor in a given mass chain. The background was determined with the ion beam stopped on a screen located about 3 m upstream from the SNC.

The fraction of neutrons due to the Rb precursor was determined in separate decay curve measurements for each mass. Decay curves were measured by multiscaling the SNC output during successive cycles of beam on and beam off. Neutron decay curves at mass 96 give only a single component due to ^{96}Rb . However at masses 97-99, there are components due to neutrons from Sr and Y precursors in addition to the Rb component. The initial activities from the decay curve analysis were converted to saturation activities to calculate the fraction of Rb neutrons in the steady state experiments.

The relevant data for determining the neutron emission rate, C, are given in Table III. Note that the Rb fraction in the first mass 98 run was quite different from the fraction in the second and third runs. The first ^{98}Rb run was taken during a different reactor cycle when TRISTAN was operating with a different ion source. The surface ionization sources for TRISTAN give different yields of Rb and Sr isobars depending on the temperature of the source and the nature of the ionizing surface. The total neutron counting rates at masses 96 and 97 were kept below 100 n/s by reducing the neutron flux on the ^{235}U target. At the same time, the background in the SNC was significantly reduced.

TABLE III. Experimental Data for Determination of Neutron Emission Rate for Rb Precursors (C)

Mass	Time (s)	Total Count Rate (n/s)	BGD Count Rate (n/s)	Fraction due to Rb (%)	C (n/s)
96	29,738	81.55	0.72 ± 0.03	100.0	80.8 ± 0.2
97	23,975	87.15	0.84 ± 0.03	98.9 ± 0.1	85.4 ± 0.2
98	20,637	10.60	2.02 ± 0.06	65.0 ± 3.0	5.6 ± 0.3
	23,749	88.29	3.83 ± 0.11	36.0 ± 3.0	30.4 ± 2.1
	65,118	11.80	2.57 ± 0.05	36.0 ± 3.0	3.3 ± 0.2
99	43,736	8.10	3.33 ± 0.05	40.0 ± 8.0	1.9 ± 0.4

An example of the time interval distribution is shown in Figure 3 for ^{98}Rb . In all cases the data over the first 200 μs were fit with two components by a least squares fitting program. The mean time of the shorter component was held fixed at the adopted residence time. The mean time of the other component was always several orders of magnitude longer. The coincident counting rate, I_s , was determined from the integral of the short component divided by the length of the run. This rate was corrected by subtracting a rate due to correlated neutrons in the background. This background was measured under a variety of conditions, i.e. reactor on or off, TRISTAN operating or not, and SNC on-line or off-line. All these background rates were the same within the statistical uncertainties. It therefore seems likely that this correlated background is due to multiple neutrons from cosmic ray showers.

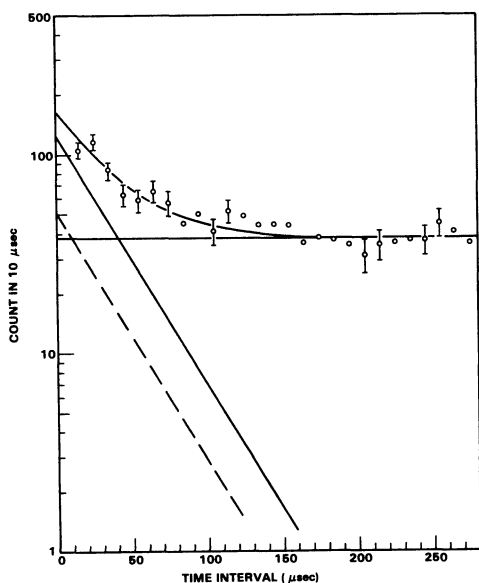


FIGURE 3. Time Interval Distribution for ^{98}Rb . Solid lines indicate best fit of two exponential components. Dashed lines show amount of correlated background as determined from low-rate ^{97}Rb experiment.

The one exception to these comments is the data associated with the first ^{98}Rb run which was taken during a different reactor cycle. The only background data available for this run is a ^{97}Rb run. Later experiments showed that the coincident neutron rate from ^{97}Rb is equal to the background rate. The coincident neutron rate for the ^{97}Rb run in question was somewhat lower than the background rates measured during a later reactor cycle. However between reactor cycles, some minor modifications to the SNC electronics were made which might account for the different background rate.

The relevant data for determining the coincident neutron rates are given in Table IV. A net positive coincidence rate was observed in three different runs for ^{98}Rb . At masses 96, 97, and 99 there was no statistically significant coincidence rate. For $^{96,97,99}\text{Rb}$ we adopt twice the statistical uncertainty as an upper limit on the coincident count rate.

3. Results

The data for C and I_s given in Tables III and IV were used in Equation 3 to calculate P_{1n}/P_{2n} as shown in Table V. The three positive results for ^{98}Rb were combined into a weighted average value of C/I_s . The P_n values were taken from a recent summary.⁹⁾ The P_{2n} for ^{98}Rb of $(0.060 \pm 0.009)\%$ is lower than the upper limit given by Roeckl, et al¹⁰⁾ of $<0.1\%$. This is the first observation of beta-delayed two-neutron emission for a nuclide in the fission product mass region. The upper limits for the P_{2n} of ^{96}Rb , ^{97}Rb , and ^{99}Rb are consistent with the small or negative energy windows predicted for this process. Our sensitivity for two-neutron detection is limited more by the correlated background than by the random singles rate.

TABLE IV. Experimental Data for Determination of Neutron Coincidence Rate (I_S)

Mass	Total Neutron Coincidence Rate (s ⁻¹)	Correlated Background Rate (s ⁻¹)	I_S (s ⁻¹)	Upper Limit ^a (I_S (s ⁻¹))
96	0.0076 ± 0.0066	0.0120 ± 0.0003	-0.0044 ± 0.0066	<0.013
97	0.0101 ± 0.0080	0.0120 ± 0.0003	-0.0020 ± 0.0080	<0.016
98	0.0212 ± 0.0019	0.0087 ± 0.0011	+0.0125 ± 0.0022	
	0.0722 ± 0.0085	0.0120 ± 0.0003	+0.0602 ± 0.0085	
	0.0197 ± 0.0009	0.0120 ± 0.0003	+0.0077 ± 0.0009	
99	0.0109 ± 0.0009	0.0120 ± 0.0003	-0.0011 ± 0.0010	<0.002

a 2σ

TABLE V. Data for Calculation of P_{2n} (or upper limit)

Mass	C/I_S	P_{1n}/P_{2n}	P_n (%) ^a	P_{2n} (%)
96	>6124	>3611	13.3 ± 0.5	<0.004
97	>5325	>3140	25.1 ± 1.3	<0.008
98	446 ± 83			
	505 ± 79			
	431 ± 59			
	455 ± 41 ^b	267 ± 33	16.1 ± 1.3	0.060 ± 0.009
99	>1005	>591	14.0 ± 3.0	<0.024

a Reference 9

b Weighted Average

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DISCUSSION

G. Rudstam: Did you analyze your decay curves to get the P_n -values for ⁹⁷Y and ⁹⁸Y?

P.L. Reeder: We have the data to obtain P_n values for Y and Sr precursors but it is not yet analyzed.