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DECAY OF THE 514-keV METASTABLE STATE IN ^{85}Rb

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An attempt was made to observe double gamma ray ($\gamma\gamma$) decay of the 1 μs metastable $9/2^+$ state in ^{85}Rb. In the measurements coincidence technique and three-parameter recording of data were applied. The upper limit $w_{\gamma\gamma}/w_\gamma \leq 1.1 \cdot 10^{-6}$, derived from the data assuming the E1M1 $\gamma\gamma$ decay, is considerably lower than the theoretical prediction based on the single-particle model and oscillator potential. From the data a new value of the branching ratio of the 363—151 keV cascade of $(1.43 \pm 0.24) \cdot 10^{-5}$ was derived, which is larger than the previous results of Alväger et al. and of Meyer et al. For the 233—281 keV cascade an upper limit of the branching ratio of $\leq 4 \cdot 10^{-7}$ was obtained.

1. Introduction

Investigation of $\gamma\gamma$ decay, i. e. of the nuclear deexcitation process by emission of a photon pair in which the two photons of the pair continuously share the transition energy, has been a challenge for over three decades. While the theory of the process, beginning with the early considerations[1], developed to the stage of giving detailed predictions for many transitions[2,3], the results of experimental investigations are rather limited. For transitions between states with nonzero angular moments so far only upper limits have been obtained[4,5]. Many investigations of the $0^+ \Rightarrow 0^+$ $\gamma\gamma$ transitions were made[6] and some achieved positive results. In a more recent investigation Schirmer et al.[7] applied a complex experimental system (a scintillation crystal-ball detector) and obtained excellent results for the $0^+ \Rightarrow 0^+$ $\gamma\gamma$ decay in ^{40}Ca and in ^{90}Zr.

Intermediate states play an important role in $\gamma\gamma$ decay. The excited states and their decay in ^{85}Rb have been studied by investigating the decay of neighbouring isobars, and by nuclear excitation and reactions[8]. Meyer et al.[9] made a detailed study of the decay schemes of ^{85m}Kr, ^{85}Kr, ^{85m}Sr and ^{85}Sr which yielded accurate data on levels of ^{85}Rb up to 1 MeV excitation. States of ^{85}Rb up to about 2.4 MeV excitation were recently investigated by Fazzini et al.[10] by proton capture in ^{84}Kr and by Coulomb excitation.

Present investigation was undertaken with the aim to observe $\gamma\gamma$ decay of the 514 keV metastable state in ^{85}Rb. In many respects it follows the methods of the previous measurement of Alväger et al.[5]. Some improvements of the measuring technique and in the analysis of data have been introduced, and data were collected for a much longer time. The experiment was designed to exceed the sensitivity required to observe the expected rate of $\gamma\gamma$ decay events estimated from the relative transition probability (about $8 \cdot 10^{-5}$), which was calculated from the results of the theory of Grechukhin[3].

2. *Measurements*

The 1 μs metastable state of ^{85}Rb was generated in the decay of ^{85}Sr (see Fig. 1).In the measurements six sources were used, which had been made from three shipments of $^{85}SrCl_2$ in a 0.5 M HCl solution (supplied by New England Nuclear, Boston, Mass., U. S. A.). Each source was made by placing a small droplet of the solution onto a polyethylene foil and by fixing the dried spot of $^{85}SrCl_2$ by cellotape. Initial strengths of the sources were 92, 163, 102, 128, 148 and 144 kBq.

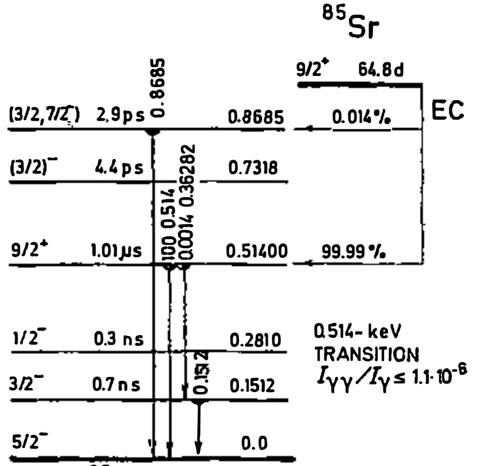

Fig. 1. Decay scheme of ^{85}Sr. New results on the decay of the 514-keV state, i. e. branching ratio of the 363-keV transition and upper limit of the relative transition probability of $\gamma\gamma$ decay, are included.

Two 38 mm diam. NaI(Tl) scintillation detectors, mounted on XP1020 photomultiplier tubes, were used for the detection of photons. The detectors were placed in lead shields and they viewed the photons emitted from the source through conical openings in a 90° geometry. In front of each detector a 1 mm thick copper absorber was placed to reduce the counting rate of the rubidium and lead KX-rays. Pulses from the anodes of the photomultipliers were lead by short cables to the discriminators (ORTEC Model 463), and were used for timing. For each coincidence event ($2\tau \approx 50$ ns) the time difference was determined by means of a time-to-amplitude converter and an analog-to-digital converter (ADC), and was recorded as the time channel (k_0). Time resolution (FWHM) of the system was about 1.5 ns. Pulses from the ninth dynodes of the photomultipliers were amplified, lead into the ADCs, and recorded as energy channels (k_1 and k_2). Coincidence rate was slow, between 30 and 110 events per hour. The recorded three-parameter data were analyzed off-line in a UNIVAC 1110 computer and in personal computers.

Absolute sensitivity of the scintillation detectors was determined by measuring the singles' counting rates with a set of calibrated sources (supplied by Radiochemical Centre, Amersham, England). The same method was applied to determine the number of counts in a peak in the analysis of calibration spectra and when analyzing the spectra derived from the three-parameter data. An interpolation procedure yielded the absolute sensitivity of the detectors in the required range of energies.

Although the measurements were made at a stable room temperature, shifts in the positions of the peaks of up to two channels were observed. In the course of the measurements some changes of the apparatus have also been made. For these reasons the recorded three-parameter data were divided into five groups, so that the spread of peak positions in each group was less than one channel. Total times of collection of data for the five groups were 1653, 625, 2004, 2204 and 1561 hours. The corresponding total numbers of 514-keV gamma rays emitted from the sources of $5.81 \cdot 10^{11}$, $3.64 \cdot 10^{11}$, $8.94 \cdot 10^{11}$, $9.83 \cdot 10^{11}$ and $7.84 \cdot 10^{11}$, respectively, were determined from the singles' counting rates of 514 keV gamma rays, taking into account the decay of the sources.

Checks of the radioactive purity of the sources were made by means of a Ge(Li) spectrometer system. A weak ^{75}Se contamination was found in two of the three shipments of ^{85}Sr.

3. *Analysis of data*

Each of the five groups of records was separately analyzed. From the three-parameter data, timing spectra were made for various sections of the $k_1 - k_2$ field with the aim to select the optimal limits of timing (k_0) channels and to determine reliable values of the coincidence efficiency. Subsequently various single-parameter (k_1 or k_2) energy spectra and two-parameter ($k_1 - k_2$) tables of numbers of events were made for energy calibration, and finally for analysis of the decay of the ^{85}Rb metastable state by emission of a pair of gamma rays. Since the analyzed timing and energy spectra were based on the complete sets of data (separately for each of the five groups), uncertainties due to small shifts in peak positions were largely eliminated. The energy calibration points were determined from the projections of the peak due to accidental coincidences of 514-keV gamma rays and of the peaks due to the 136—265, 121—280 and 97—304 keV cascades in the weak (about 0.02%) ^{75}Se contamination of the sources. Positions and widths of the peaks in the k_1 and k_2 spectra were determined using a non-linear least-squares routine. The fitting functions were defined numerically as »line profiles« which were derived from the singles' spectra. Interpolation was made using a SPLINE routine.

Decay of the 514 keV metastable state to the ground state of ^{85}Rb by emission of two gamma rays was expected to yield data on the $E_1 + E_2 = 514$ keV line in the $k_1 - k_2$ field. One of the major concerns was, therefore, to reduce background in this region to a low value. Main cause of the background were coincidences of pulses due to accidental simultaneous incidences of a 514 keV gamma ray into one detector and of another 514 keV gamma ray into the other detector, either

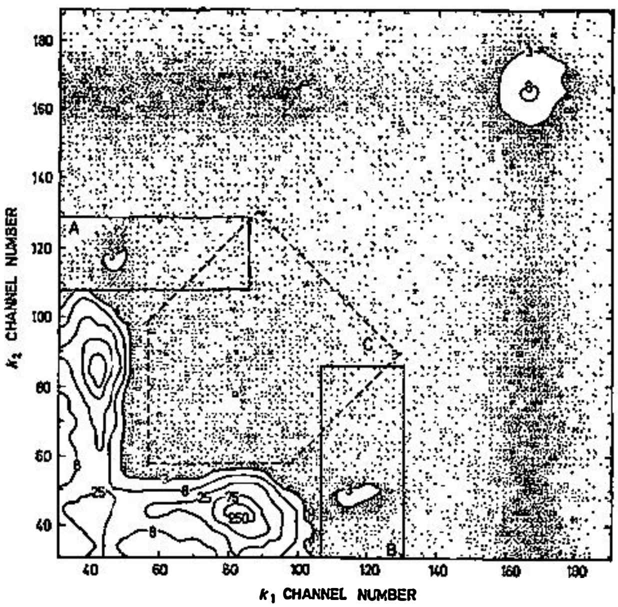

Fig. 2. Illustration of the $k_1 - k_2$ field of numbers of events made from the third group of data. Data are shown by dots when the number of events per 2 × 2 squared channels is less than 11, while for larger numbers data are indicated by the equal-density lines (densities per squared channel are given on the lines). The peak at $(k_1, k_2) \approx (166, 166)$ is due to accidentalcoinciddences of pulses from total-energy absorption of a pair of 514-keV gamma rays in the two detectors (the »514 — 514 keV« peak). Increased density of dots in the bands extending from the 514—514 keV peak, parallel to the k_1 or k_2 axis is due to accidental coincidences of pulses from the total-energy absorption of 514-keV gamma rays in one and their partial absorption (»Compton continuum«) in the other detector. The peaks at $(k_1, k_2) \approx (42, 87)$ and (87, 43) are due to the real coincidences of the cascade transitions in ^{75}Se. The pair of »weak« peaks at $(k_1, k_2) \approx (47, 120)$ and (118, 49) is due to the (363 — 151) cascade in ^{85}Rb. Projections of data in the cuts A and B onto the k_1, and k_2 axes, respectively (Fig. 3), were used to determine the intensity of the 363 — 151 keV cascade. Sum spectra were obtained by projecting the data in cuts indicated by C onto the $k_1 + k_2$ axis, and were used to determine upperlimit of the relative $\gamma\gamma$ decay transition probability.

of the two being partially absorbed (the accidental Compton continuum — Compton continuum events). To keep the background low weak sources had to be used.

On the $E_1 + E_2 = 514$ keV line two weak, symmetrically positioned peaks were observed (see Fig. 2). Identification of the peaks as due to the 363—151 keV cascade in ^{85}Rb is based on the energy and on the decay arguments. From the five groups of data, the curve fitting, and the energy calibration, ten values of energy of the cascade gamma rays, E_1 and E_2, were derived, the average values being $E_1 = (150.6 \pm 1.3)$ keV and $E_2 = (362.6 \pm 1.8)$ keV, in good agreement with the known values[9] 151.18 and 362.8 keV, respectively. For each of the six sources used in the measurements the numbers of counts in the weak peaks allowed the determination of the decay constant. Average value of the results for the half-life thus determined was (67 ± 4) days, in good agreement with the known value for ^{85}Sr of 64.8 days.

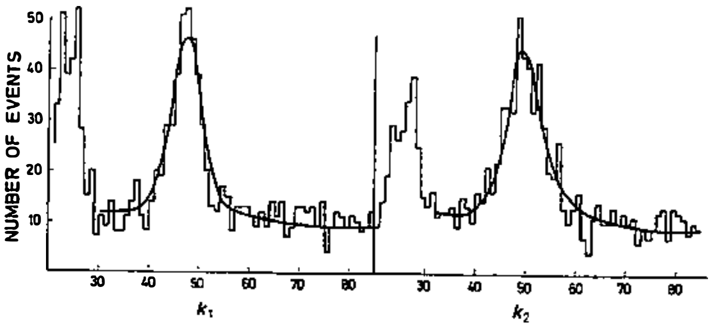

Fig. 3. Projections of data in the cuts A and B shown in Fig. 2 onto the k_1 and k_2 axes, respectively. Analyses of the spectra yielded the full curves shown in the figures. The peaks at left are due to the lead KX-rays.

A careful check of the $k_1 - k_2$ tables of numbers of events was made for peaks due to contamination which might have contributed to the counting in the two »weak« peaks. Also a detailed inspection of the Table of Isotopes[8] was made. On the basis of the above considerations the counts in the two weak peaks are considered to be due to the 363—151 keV cascade in ^{85}Rb. They were used to determine the branching ratio of the 363—151 cascade. For each of the five groups of data two-parameter tables of numbers of events (an illustration is shown in Fig. 2) were made. Cuts in the tables around the weak peaks and their projections onto k_1 or k_2 axis (see Fig. 3) yielded spectra with pronounced peaks due to the 363—151 keV cascade. A standard least-squares analysis was applied to obtain the numbers of counts. The fitting functions were defined numerically, as described above.

A continuous ridge due to $\gamma\gamma$ decay was expected along the line $E_1 + E_2 = 514$ keV, crossing the two weak peaks. The sum-energy spectra were made for cuts in the two-parameter tables of numbers of events in the region between the two weak peaks. The boundary of a cut is shown in Fig. 2 by the dashed line (C). Each of the five sum spectra was analysed separately. Also a joint sum spectrum was made by adding the five sum spectra after a proper adjustment of the $k_1 + k_2$

scales were made (Fig. 4). Prior to adding the spectra, corrections were also made for reduced numbers of channels which were added when making each of the five sum spectra (due to the taper of boundaries C for lower values of $k_1 + k_2$). In the analysis of the sum spectra the function representing the background due to Compton continuum — Compton continuum accidental coincidence events was calculated from the singles' spectra due to the 514-keV gamma rays. The width of the line due to $\gamma\gamma$ decay was (for each sum spectrum) assumed to be equal to the width of the weak peaks projected onto the $k_1 + k_2$ axis.

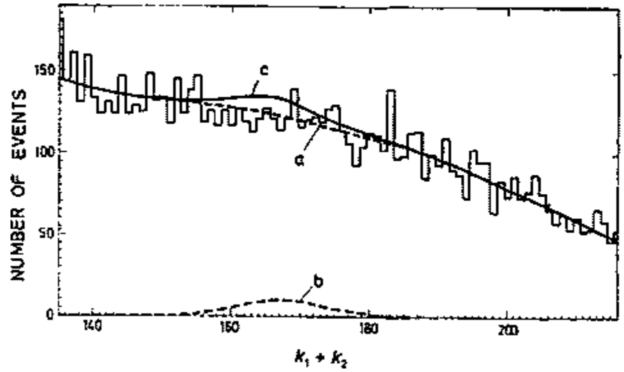

Fig. 4. Histogram shows the sum spectrum obtained by adding five sum spectra, after the energy scales were adjusted and the numbers of counts at lower ends of the spectra corrected for taper of the boundary C (Fig. 2). The curves shown the results of least-squares fit: (a) »Compton continuum — Compton continuum« sum spectrum, (b) the peak which would have been due to 131 detected $\gamma\gamma$ events, and (c) sum of the two curves.

Analyses of the two-parameter tables of numbers of events were also made. The fitting function was composed of four two-dimensional functions representing (i) Compton continuum — Compton continuum distribution due to accidental coincidences, (ii) a pair of peaks due to the real coincidences of the 363—151 keV cascade transitions, (iii) a pair of peaks due to the assumed 281—233 keV gamma-ray cascade, and (iv) the ridge along the line $E_1 + E_2 = 514$ keV due to the assumed $\gamma\gamma$ decay. The ridge due to $\gamma\gamma$ decay was assumed to have a Gaussian profile, while all other functions were constructed from the real singles' spectra and used as numerically defined functions (as in the energy-calibration fits).

Non-linear least-squares fits width equal weights were made to obtain the numbers of events. The same fits were repeated with weights equal to the reciprocals of the numbers of counts [taking 1 for zero count[11]] to obtain the corresponding statistical errors.

4. Results and discussion

For each of the five groups of data the analysis of the single-parameter spectra, obtained by projecting the numbers of events in sections indicated by A, B and C in Fig. 2 onto the k_1, the k_2, and the $k_1 + k_2$ axis, respectively, and of the two-parameter spectra, i. e. of the sections of the $k_1 - k_2$ field, were made. The results of the two methods of analysis are in good agreement. The analysis of single-parameter spectra seems to be more reliable, i. e. less sensitive to e. g. the choice of the fitting functions.

The sum spectra were made to accept $\gamma\gamma$ events in the energy range $E_a = 184$ to $E_b = 328$ keV for each photon of a $\gamma\gamma$ pair. For that energy range the least-squares analysis of the five sum spectra yielded the following numbers of counts due to $\gamma\gamma$ decay: -25 ± 33, -18 ± 31, 16 ± 39, -38 ± 40 and 16 ± 39, the total result being -49 ± 82 events. The analysis of the joint spectrum (shown in Fig. 4) yielded -40 ± 67 events. The upper limit on the number of events due to $\gamma\gamma$ decay in all five groups of data at the 95% confidence level

$$n_{\gamma\gamma} \leq 131$$

was calculated (assuming a null result) from the errors.

Ratio of the total transition probability of $\gamma\gamma$ decay and of the direct gamma-ray emission transition probability is given by

$$\frac{w_{\gamma\gamma}}{w_\gamma} = \frac{n_{\gamma\gamma}}{n_\gamma F}$$

where $n_{\gamma\gamma}$ is the number of detected $\gamma\gamma$ decay events, n_γ the number of direct gamma-ray emissions, and F the efficiency of the system for the detection of $\gamma\gamma$-decay events. To a good approximation F can be factorized as follows,

$$F = \varepsilon_c \overline{w(\Theta)}\, I$$

where ε_c is the coincidence efficiency.

$$\overline{w(\Theta)} = (1 + a_2 \overline{\cos^2 \Theta})/(1 + a_2/3)$$

is the averaged normalized angular distribution, and

$$I = \frac{\int_{E_a}^{E_b} f(E) \cdot [\Omega_1 \varepsilon_1 (E) \Omega_2 \varepsilon_2 (E_0 - E) + \Omega_1 \varepsilon_1 (E_0 - E) \Omega_2 \varepsilon_2 (E)]\, \mathrm{d}E}{(4\pi)^2 \int_0^{E_0} f(E)\, \mathrm{d}E}$$

is the efficiency of detection which takes into account the energy interval E_a to E_b accepted in determining $n_{\gamma\gamma}$, absorption, and the solid angles of the detectors. In the expression for I the solid angles of the detectors are denoted by Ω_1 and Ω_2, the peak efficiencies by ε_1 and ε_2, and the energy distribution of $\gamma\gamma$ decay by $f(E)$.

During the collection of the data of the five groups the total number of emitted 514-keV gamma rays was $3.6 \cdot 10^{12}$. In the analysis only E1M1 (or M1E1) $\gamma\gamma$ decay was assumed because higher combinations of multipoles have a much smaller probability. Because the initial state is a $9/2^+$ and the ground state a $5/2^-$ state, the virtual intermediate state was assumed to be a $7/2^+$ or $7/2^-$ state. For such a sequence of states $a_2 = 1/13$. The value of $\cos^2 \Theta = 0.02$ was calculated by numerical integration. The integral in the denominator of the expression for I was also calculated by numerical integration, assuming the energy distribution

$$f(E) = \text{const}\ E^3 (E_0 - E)^3,$$

and the interpolated values of the peak efficiencies. The result for the detection efficiency of the system is $I = 0.026 \cdot 10^{-6}$. The coincidence efficiency of the data collection was $\varepsilon_c = 0.97$. Using the quoted values the upper limit

$$w_{\gamma\gamma}/w_\gamma \leq 1.1 \cdot 10^{-6}$$

was deduced.

Grechukhin[3] gives detailed expressions for the calculation of the theoretical estimates of the relative $\gamma\gamma$ transition probabilities. From his results, assuming only one intermediate state, the $7/2^-$ excited state in ^{85}Rb at 868.6 keV[9,10], one obtains

$$w_{\gamma\gamma}/w_\gamma = 8.1 \cdot 10^{-6}.$$

Other $7/2^+$ or $7/2^-$ excited states in ^{85}Rb have not been found below an excitation of 1.4 MeV [one at 1.445, one at 1.930 and one at 2.028 MeV possibly have angular momentum 7/2[10]]. Because the contribution to the transition amplitude strongly decreases as the excitation energy increases, contributions of higher 7/2 states are expected to be small. We, therefore, conclude that the single-particle model with the oscillator potential, in the case of the 514-keV metastable state in ^{85}Rb, overestimates the relative transition probability of the $\gamma\gamma$ decay by a factor of more than seven.

The relative intensity of the 363—151 keV gamma-ray cascade was determined separately for each of the two weak peaks for each of the five groups of data. The average value of the ten results is

$$\frac{w(\gamma(363) - \gamma(151))}{w(\gamma(514))} = (1.41 \pm 0.26) \cdot 10^{-6}.$$

This result is by about a factor of two larger than the result $(6 \pm 3) \cdot 10^{-6}$ of Alväger et al.[5] obtained in a coincidence measurement also with ^{85}Sr sources, and the result $(5 \pm 1) \cdot 10^{-6}$ of Meyer et al.[9] derived from the singles' spectra in the decay of ^{85}Kr.

To deduce the branching ratio of the 363-keV transition electron conversion has to be taken into account. For the 514-keV transition $\alpha = 0.0066$[8]. Conversion coefficients of the 363- and 151-keV transitions have not been experimentally determined, but reliable theoretical values can be obtained. From the tables of Hager and Seltzer[12] one finds $\alpha(Z = 37, \text{E3}, 363\ \text{keV}) = 0.033$ and $\alpha(Z = 37,$

M1, 151 keV) = 0.047. Our result for the branching ratio of the 363-keV transitions is, therefore,

$$\frac{w\,(363\ \text{keV})}{w\,(514\ \text{keV}) + w\,(363\ \text{keV})} = (1.43 \pm 0.26) \cdot 10^{-5}.$$

From the results of the analyses of the sections of the $k_1 - k_2$ fields the upper limit of the relative intensity of the 281—233 keV cascade

$$\frac{w\,(281 - 233\,\text{keV})}{w\,(514\ \text{keV})} \leq 4 \cdot 10^{-7}$$

was derived. According to the single-particle estimates this cascade is expected to be several orders of magnitude weaker than the present limit.

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RASPAD 514-keV METASTABILNOG STANJA U ^{85}Rb

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Načinjen je pokušaj da se nađe dvojni gama ($\gamma\gamma$) raspad 1 μs metastabilnog $9/2^+$ stanja u ^{85}Rb. U mjerenjima primijenjena je koincidentna metoda i troparametarsko bilježenje podataka. Gornja granica, $w_{\gamma\gamma}/w_\gamma \leq 1.1 \cdot 10^{-6}$, izvedena iz podataka pretpostavljajući E1M1 $\gamma\gamma$ raspad, znatno je niža od teorijske vrijednosti izračunate na osnovi Grechuhinove teorije koja se zasniva na jednočestičnom modelu s oscilatornim potencijalom. Na osnovi mjernih podataka dobivena je nova vrijednost omjera grananja za 363 — 151 keV kaskadni raspad stanja na 514 keV od $(1.43 \pm 0.24) \cdot 10^{-5}$, koji je veći od ranijih rezultata Alvägera i sur. i Meyera i sur. Za kaskadu 233—281 keV određena je gornja granica omjera grananja od $\leq 4 \cdot 10^{-7}$.