A NEW BETA DELAYED PROTON EMITTER ^{93m}Ru

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Abstract

The 10.8 s, 1/2$^-$ isomer of ^{93}Ru is shown to be a β-delayed proton precursor. The absolute branching ratio for this decay mode is determined to be $(2.7 \pm 0.5)\cdot 10^{-4}$. The measured total decay energy of ^{93m}Ru is 7.06 ± 0.20 MeV. The delayed proton spectrum has prominent peaks at 2.481, 2.534 and 2.557 MeV indicating structural effects in the β-strength function. The level structure of ^{93}Tc was also studied by the ^{92}Mo(p,γ)^{93}Tc resonance reaction in the proton energy range 2.00 MeV < E_p < 2.68 MeV. Individual resonances were well resolved with an energy resolution of 1 keV.

1. Introduction

Studies of β-delayed charged particle emission have yielded a wealth of information on statistical properties of nuclides far from the region of β-stability, as discussed in detail in several review papers $^{1-4}$). We have recently carried out a study on delayed particle emission of a new nuclide ^{59}Zn (ref. 5) and subsequently extended our investigation to the heavier nuclides ^{65}Ge and ^{93}Ru. These three nuclides, which cover a relatively broad range in mass, are the lightest known proton precursor nuclides in the T_Z = -1/2, +1/2 and +5/2 series, respectively, and can all be produced by (^{3}He, 2n) reactions. All other precursor nuclides with Z ≥ 28 and within reach of the (^{3}He, 2n) reaction have a less favourable energy window for delayed proton emission. The three mentioned precursors are therefore unique in the sense that the high resolution characteristic of (p,γ) and (p,p_0) resonance reactions can be used for detailed study of the level properties of the emitter nuclei in the energy region corresponding to delayed proton emission. Valuable spectroscopic information on proton unbound levels may also be available through other types of reactions, such as (^{3}He, d) stripping reactions ^{5}).

In this report we deal with the new delayed proton precursor nuclide, ^{93}Ru, in which delayed proton emission is associated with the $J^\pi = 1/2^-$, $T_{1/2}$ = 10.8 s isomeric state at an excitation of 0.734 MeV. The decay properties of ^{93m}Ru have been studied by de Lange et al. ^{6}). The predicted EC decay energy of ^{93g}Ru is 6.3 MeV and the proton binding energy of ^{93}Tc is 4091.6 ± 2.9 keV ^{7}). The emitter nuclide ^{93}Tc has a closed N = 50 neutron shell and only the $J^\pi = 1/2^-$ and $3/2^-$ final states are populated in the allowed β-decay. These two facts suggest that the density of final states available for allowed β-decay of ^{93m}Ru should be comparatively low. It is therefore of interest whether the peaks in the proton spectra can be interpreted in terms of structural effects related to individual transitions, or in terms of Porter - Thomas fluctuations in the transition probabilities. For this part of our work we have taken advantage of the high resolution available in (p,γ) resonance reaction studies. Useful information on low-lying, low-spin bound states of ^{93}Tc have been deduced from average resonance spectroscopy ^{8}), but no previous high resolution (p,γ) studies of resonance levels in ^{93}Tc have been performed.

2. Experimental

2.1. Beta decay and beta-delayed protons

Neutron deficient ^{93}Ru nuclides were produced by bombarding isotopically enriched 0.5 mg/cm^2 ^{92}Mo targets with a beam of 24 MeV ^{3}He ions from the University of Jyväskylä MC-20 and the University of Oslo MC-35 cyclotrons. Targets were prepared by evaporating 98.21% enriched ^{92}MoO$_3$ on 0.17 mg/cm^2 thick Al backing foils. Recoils ejected from the stack of four adjacent target foils were thermalized in 1.5 atm helium and then transported with NaCl-loaded helium through a 3 or 8 m long and 0.8 mm diameter capillary to a tape transport device. The activity was collected onto an aluminized mylar tape from the emerging gas jet and was moved according to a preselected time cycle to the detection position incorporating detectors for beta-, gamma- and delayed particle counting ^{9}). Radioactive sources collected by the present technique are practically massless and are thus ideally suited for high resolution delayed particle spectroscopy.

A statistical-model calculation predicts that the beta-delayed proton spectrum of ^{93m}Ru lies in the proton energy interval from 1.5 to 3.0 MeV. In the detection of these protons both a detector telescope and a high resolution single counter were used. The telescope consisted of a 31 μm, 50 mm^2 fully depleted ΔE and 100 μm, 300 mm^2 partially depleted E Si(Au) detectors. In the high resolution experiments the sensitive volume of the 100 μm detector was decreased by lowering the bias voltage from its nominal value of 105 V in order to reduce the intense background caused by the multiple beta scattering. In the telescope operation linear signals from both detectors were stored event-by-event on a magnetic tape for later analysis. To determine the half-life associated with beta-delayed protons, the time elapsed from the end of the tape transport cycle was also recorded for each event. Calibration of the proton counters was accomplished by detecting the beta-delayed proton emitters ^{25}Si ^{10,11}) and ^{40}Sc^{10,12}), produced in high yields in the ^{24}Mg(^{3}He,2n) and ^{40}Ca(p,n) reactions, respectively. The energy resolution of the telescope and the single counter were 50 and 20 keV, respectively.

Simultaneously with the delayed proton counting, gamma rays associated with the beta decay were recorded with a 70 cm^3 Ge(Li) detector positioned in a face-to-face geometry with the proton counter. This allowed determination of the absolute proton branching through comparison of the intensities of the known gamma rays and observed protons. The relative efficiencies of the particle counter and the Ge(Li)-detector were determined by the known intensity ratio, 7.87 ± 0.12, of the alpha particles, and the 351 keV gamma-ray following the alpha decay of ^{211}Bi [13]).

The beta end point energy of ^{93}Ru was measured by a solid state telescope operating in coincidence mode. It consisted of a 3 mm thick Si(Li) ΔE detector with a sensitive area of 2 cm^2 and a pure germanium E detector, 5 mm thick and 10 mm in diameter. The pulses from the two detectors are summed to give the total energy deposited in the system. The resolution of this spectrometer was 25 keV, reflecting mainly the effect of the 29 mg/cm^2 dead layer in the Si(Li) detector. Calibrations of the ΔE and E detectors were made separately using the photopeaks and Compton edges of well known gamma rays. The operation of the telescope was also checked using positron emitters with well known end-point energies between 3.0 and 7.3 MeV. Again, the linear signals from the detectors were stored event-by-event on a magnetic tape for subsequent analysis. A more detailed description of this device will be given elsewhere [14]).

2.2. Proton capture experiments

Proton capture reaction studies were performed at the Helsinki University 2.5 MV van de Graaff accelerator. Metallic molybdenum powder enriched to 94.1% in ^{92}Mo was pressed into a small pellet and vacuum evaporated onto a 0.4 mm Ta backing foil. The thickness of the evaporated Mo layer was about 15 nm, or 1 keV for 2.0 MeV protons. The backings were cooled by liquid nitrogen or by water to prevent the target from being melted by the beam currents which were 5-10 μA. The energy spread of the proton beam was about 1 keV. The targets were analyzed by backscattering and PIXE techniques to ensure that no interference from impurities might occur.

A relative yield curve for the $^{92}\mathrm{Mo}(p,\gamma)^{93}\mathrm{Tc}$ reaction was measured in the proton energy range $2.00\ \mathrm{MeV} < E_p < 2.68\ \mathrm{MeV}$. In the measurement the gamma ray energy window was set at $3.0\ \mathrm{MeV} < E_\gamma < 7.0\ \mathrm{MeV}$. The gamma rays were measured with a 10.2 cm diameter×12.7 cm NaI(Tl) detector at 55° at a distance of 2 cm from the target. The known resonances at $E_p = 1799.5 \pm 0.09$ keV and $E_p = 2045.3 \pm 0.8$ keV in the $^{27}\mathrm{Al}(p,\gamma)^{28}\mathrm{Si}$ reaction were used for energy calibration [10]). The decay properties of individual resonances were investigated using a 110 cm^3 Ge(Li) detector having a resolution of 3.0 keV (FWHM) at 2.614 MeV. The detector was placed at 0° relative to the beam direction and at a distance of 3 cm from the target. The thick target measurements were carried out in the same detector geometry.

3. Results

3.1. Beta-delayed proton decay of ^{93m}Ru

The energy spectrum of beta-delayed protons arising from the decay of ^{93m}Ru, as measured with the detector telescope, is shown in fig. 1(a). The tape collection and measuring periods were 20 s with a transport time of 0.3 s to the detector position. The time sequential proton spectra yield a half-life of 10 ± 2 s, in good agreement with a previously measured value of 10.8 ± 0.3 s based on a gamma decay study of ^{93m}Ru [6]). In view of the limited ^{3}He beam intensity at the Jyväskylä MC-20 Cyclotron an additional high resolution single counter experiment was performed at the MC-35 Cyclotron at the University of Oslo. The spectrum resulting from this experiment (fig. 1(b)) reflects the same characteristic features as the spectrum obtained with the telescope. There is a strong peak at 2481 ± 5 keV and several smaller intensity peaks distributed from ∿1.5 to 3.0 MeV. The total decay branching to proton emitting states was determined by comparing the total number of protons with the intensity of the 1.396 MeV gamma-ray peak [6]) in the same experimental conditions as for the ^{211}Bi efficiency calibration. From this measurement a proton branching ratio of $(2.7 \pm 0.5)\cdot 10^{-4}$ was obtained for ^{93m}Ru.

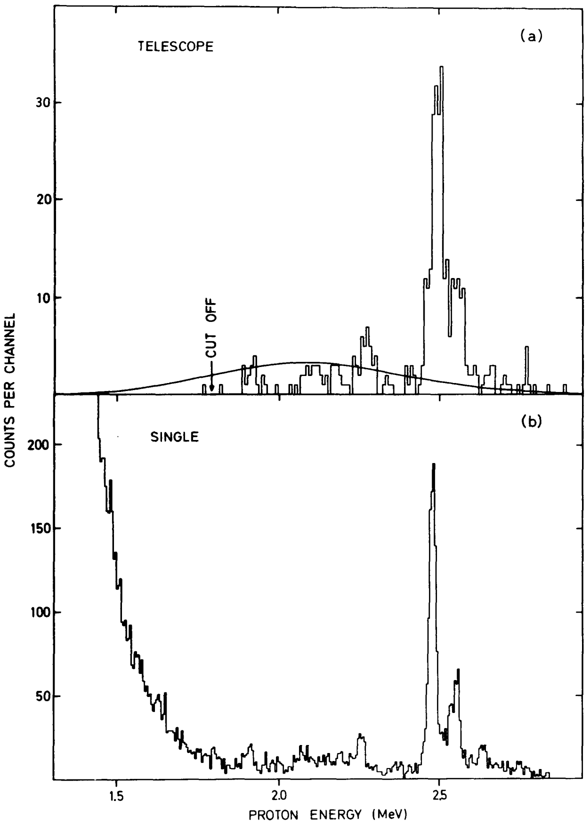

Fig. 1. The beta-delayed spectra of ^{93m}Ru obtained with (a) a telescope consisting of a 31 μm ΔE and a 100 μm E detector (FWHM 50 keV) and (b) a single 100 μm detector operating with a bias voltage of 20 V (FWHM 20 keV). The arrow in the top figure refers to cut-off energy of the telescope. The collection and measusing period was 20 s; the total integrated ^{3}He beam currents were 15 mC in the telescope run and 130 mC in the single counter experiment. The solid curve in fig. (a) shows the average proton intensity distribution calculated with a simple statistical model.

The assignment of the observed proton groups to the decay of the 1/2$^-$ isomeric state of ^{93}Ru was based on the measured half-life and the excitation function of the $^{92}\mathrm{Mo}(^3\mathrm{He},2n)^{93m}\mathrm{Ru}$ reaction monitored by the 734.4 keV isomeric transition. Moreover, the simultaneously recorded γ-ray spectrum did not provide evidence for any unknown activity with a half-life similar to the one of the observed protons. Additional support can also be drawn from comparison of the observed spectrum with the average proton intensity distribution calculated from a simple statistical model (see fig. 1(a)). Possible interference from beta-delayed proton decay of the ^{93}Ru ground state is inhibited by the very low beta feeding rate as well as by higher ℓ-values involved in the proton decay.

Statistical calculations of the average shapes of delayed particle spectra have shown remarkable similarities to observed intensity distributions[15,16]. To find the effective energy window for the delayed protons, a calculation (fig. 1(a)) along the lines of ref. 17 was performed in the present case. The total decay energy of 7060 ± 200 keV, as determined in the present work, was used for ^{93m}Ru. The level densities of the $1/2^-$ and $3/2^-$ states in ^{93}Tc populated by the allowed beta decay of ^{93m}Ru were calculated from the formula of Gilbert and Cameron[18]. The value of the level density parameter a was taken to be 9.2 MeV^{-1}, as recommended by Truran et al. [19]. The proton decay widths were calculated assuming only one final state, i.e. the 0^+ state of ^{92}Mo. Transmission coefficients for $\ell=1$ protons were obtained through the relation $T_{\ell j}(E_p) = \gamma^2_{\ell j} P_\ell(E_p)$ with $\gamma^2_{\ell j}$ = constant and $P_\ell(E_p)$ equal to the barrier penetrability[11,15]. Finally, the average E1 radiative widths of the levels under consideration were calculated from the expression given in ref. 17. In the extraction of the beta intensity distribution, a constant β-strength was assumed. The value of the statistical rate function was taken from the tabulation of Gove and Martin [20]. Although the resulting proton intensity envelope does not very well follow the experimentally observed distribution with strong intensity fluctuations, the calculation does give an energy window for protons which agrees well with the observed one and thus support the assignment of these protons to the decay of ^{93m}Ru. The calculated total proton branching is 1.1×10^{-4} , which is about one half of the observed value. Such disagreement may easily result from the many approximations implicit in the calculation.

3.2. Decay energy of ^{93}Ru

The total decay energy, Q_{EC}, of the $1/2^-$ isomeric state was deduced from the measurement of the Q_{EC} value of the $9/2^+$ ground state of ^{93}Ru. Fig. 2 shows the singles positron spectrum measured from a metallic 5 mg/cm^2 ^{92}Mo target (enriched to 94.1%) using a pulsed 24 MeV ^{3}He beam and the solid state telescope. The beam-on period was 120 s and the beta-counting was performed during the 120 s beam-off period, starting after a 30 s delay to minimize the effect of positrons from the decay of the isomeric state. The high energy part of the spectrum was assigned to the decay of ^{93g}Ru. Considering target impurities and timing, it is unlikely that any other high energy, beta-emitter could be produced. The second highest beta end-point energy of nuclides produced in the target was 4.2 MeV, and belonged to the 4.4 min decay of ^{92}Tc[13]. A Fermi-Kurie analysis of the beta-spectrum resulted in an end-point of 5300 ± 200 keV. The operation of the telescope was checked with the beta-emitters[13] ^{66}Ga (4153 ± 3 keV), ^{64}Ga (6143 ± 8 keV) and ^{54}Co (7719.8 ± 1.8 keV). Reasonable agreement with these values was obtained. The fairly large uncertainty of 200 keV for the ^{93g}Ru decay energy is mainly due to the difficulty of defining the proper fitting region in the FK-plot. The experimental result agrees quite well with the predicted value of 6300 keV given in the 1977 Mass Tables [7]. The total decay energy of ^{93m}Ru, as derived from the measured end-point of ^{93g}Ru, is thus 7060 ± 200 keV.

3.3. (p,γ) resonance structure

In addition to measuring the relative yield curve for the ^{92}Mo(p,γ)^{93}Tc reaction we investigated γ-decay patterns associated with some of the prominent peaks in the yield curve. The observed patterns involved a varying number of γ-transitions leading from the resoanace to known low lying levels in ^{93}Tc and from these levels down to the $J^\pi = 1/2^-$ isomeric state and the $J^\pi = 9/2^+$ ground state. In the proton energy region overlapping the observed intense 2481 keV peak in the delayed proton spectrum shown in fig. 1(b), the behaviour of the observed γ-ray pattern was studied as a function of energy. In the energy range 2533 keV < E_p < 2544 keV we observed two distinct γ-decay patterns, one centered at 2537 keV and the other at 2539.5 keV. The proton energy was varied in steps of 0.6 keV and singles γ-ray spectra were measured at each energy. The ratios of the most intense γ-transitions associated with the two patterns remained the same as a function of proton energy, but the yields varied in the same way as in the measurement of the yield curve in single channel mode. The 2517 keV peak in the yield curve was studied in the same fashion and again we observed the decay pattern to remain the same as we moved over the peak, varying the proton energy in steps of 0.9 keV. The observed correlation of the whole γ-decay pattern with proton energy indicates that the peaks observed in the yield curve are due to isolated resonances.

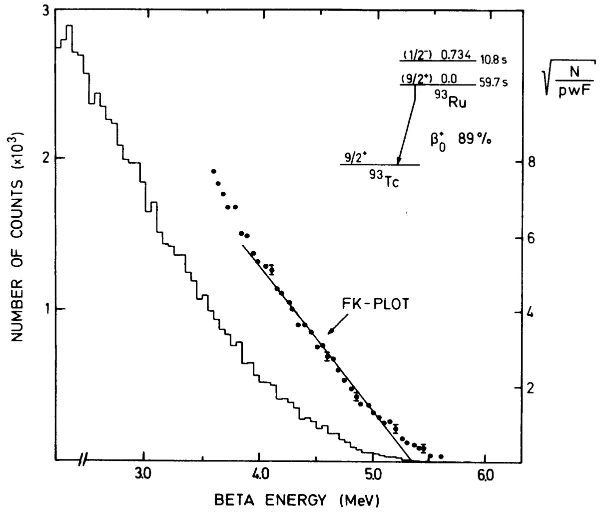

Fig. 2. Measured positron spectrum of ^{93g}Ru and its Fermi-Kurie (FK) analysis.

The predominance of γ_1-transitions leading from resonance states in the compound nucleus to the first excited state, the $J^\pi = 1/2^-$ isomeric state, is evident in the study of average resonance spectroscopy in ^{93}Tc by Close and Bearse [8]. This particular feature of the decay of resonance levels excited in ^{93}Tc by the (p,γ) reaction was also observed in our bombardment of a thick ^{92}Mo target by 2.59 MeV protons. No γ_0-transitions were observed and the γ_1-transitions were found to dominate the spectrum completely in the energy region 5.0 MeV < E_γ < 6.3 MeV; a section of the measured spectrum in this energy region is presented in fig. 3 together with a matching section of the measured yield curve of the (p,γ) reaction. All the observed γ-peaks result from γ_1-transitions and can be correlated by energy with a definite peak in the yield curve. The widths of the γ-ray peaks are compatible with those observed for single peaks in this energy region. The observed density of resonance levels strongly depopulated by γ_1-transitions is about 50 MeV^{-1}, while the number of observed resonance peaks in the yield curve is approximately 180 MeV^{-1}.

The decay scheme of ^{93m}Ru based on the results of this work and the γ-spectroscopic study by de Lange et al. [6] is shown in Fig. 4. On the left

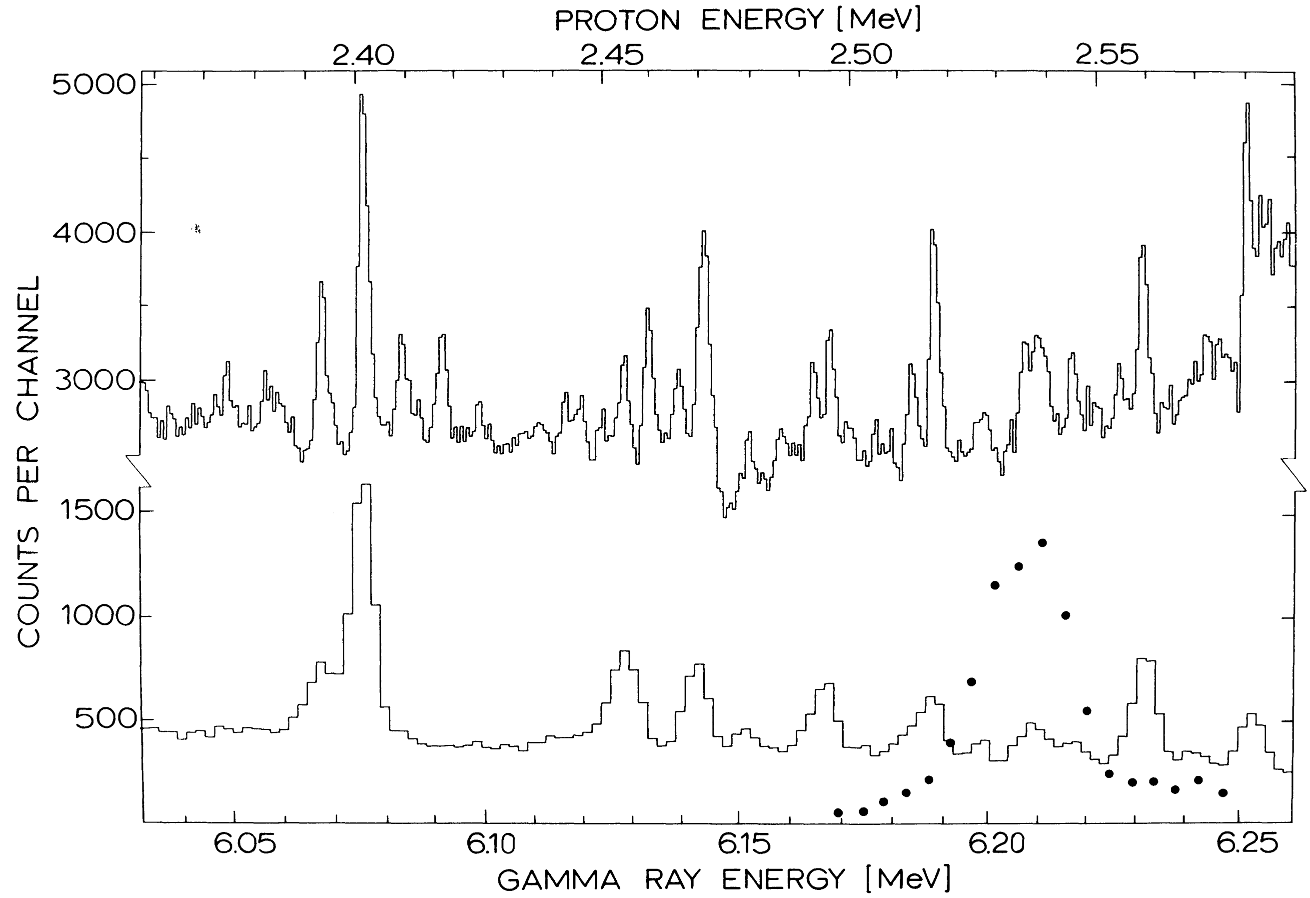

Fig. 3. The relative yield curve of the ^{92}Mo(p,γ)^{93}Tc reaction (upper spectrum) is presented with a gamma ray spectrum resulting from bombardment of a thick ^{92}Mo target with 2.59 MeV protons. The energy scales have been adjusted so that the proton energy E_p of each resonance coincides with the energy of the corresponding γ_1-transition. The most prominent peak in the measured delayed proton spectrum (fig. 1) is displayed by large dots for comparison.

hand side we present the observed γ-ray pattern of the 6601 keV level associated with the resonance at E_p = 2537 keV in the ^{92}Mo(p,γ)^{93}Tc reaction. The (2481 ± 5) keV delayed proton peak corresponding to E_X = (6600 ± 6) keV, is energetically most closely associated with this resonance. However, taking into account the uncertainty in the peak location, it could also be associated with an adjacent resonance at 2539.5 keV, or possibly with some unobserved weaker resonance. Because of a direct γ-transition from the 6601 resonance level to the 1194 keV level with $J^\pi = 5/2^+$ [6,8], the spin assignment $J^\pi = 1/2^-$ for the resonance level can be excluded. Therefore, if the 2481 keV delayed proton group and the 6601 resonance are associated with the same level, only a $J^\pi = 3/2^-$ assignment is compatible with the allowed character of the feeding β-transition.

4. Discussion

4.1. Level density

For the emitter nucleus ^{93}Tc a value for the level density parameter can be deduced from the observed level spacing D = 2.0 ± 0.4 keV in the neighbourhood of the $1/2^+$ analog resonance at E_x = 9.34 MeV [21]. With this experimental constraint Gilbert Cameron's level density formula [18] yields the value a = 9.0 ± 0.2 MeV^{-1}, which is close to the value a = 9.2 MeV^{-1} predicted by Truran and Cameron[19]. With a = 9.0 MeV^{-1} Gilbert Cameron's formula predicts a level density of about 40 MeV^{-1} for J = 1/2 states of one parity at E_x = 6.6 MeV. The density of levels with $J^\pi = 1/2^-$ and $3/2^-$, both of which are available to allowed β-decay from initial level $J = 1/2^-$ in ^{93}Ru, is 120 MeV^{-1} according to the formula. The observed density of resonance peaks in the (p,γ) reaction yield curve at E_x = 6.6 MeV is about 200 MeV^{-1}. This value is in qualitative agreement with the predicted density of levels, because only low ℓ values contribute to the observed resonances, and fluctuations in level widths and level spacings result in a number of missing levels. In conclusion, it is apparent that, with the high energy resolution associated with the (p,γ) resonance experiments, it is possible to study individual resonance states in ^{93}Tc.

Delayed proton emission of ^{99}Cd, which is near to a double closed shell (N = Z = 50), has been studied in detail [22]. Because of several common features, a comparison of the delayed proton emission of ^{99}Cd and ^{93}Ru is of interest. A level density value a = 8.5 MeV^{-1} was deduced for the emitter nuclide ^{99}Ag by a statistical model analysis of the delayed proton spectra. The predicted proton binding energy of ^{99}Ag is 2.54 MeV and the Q_{EC} value is 7.36 MeV [23]. For delayed protons of 2.5 MeV, corresponding to an excitation of about 5 MeV, the predicted density of J = 5/2 levels, using a = 8.5 MeV^{-1}, is 25 MeV^{-1}. In the observed delayed proton spectrum of ^{99}Cd large intensity fluctuations are observed, but the location of this nuclide far from stability does not presently allow a detailed spectroscopic study of the nature of these fluctuations.

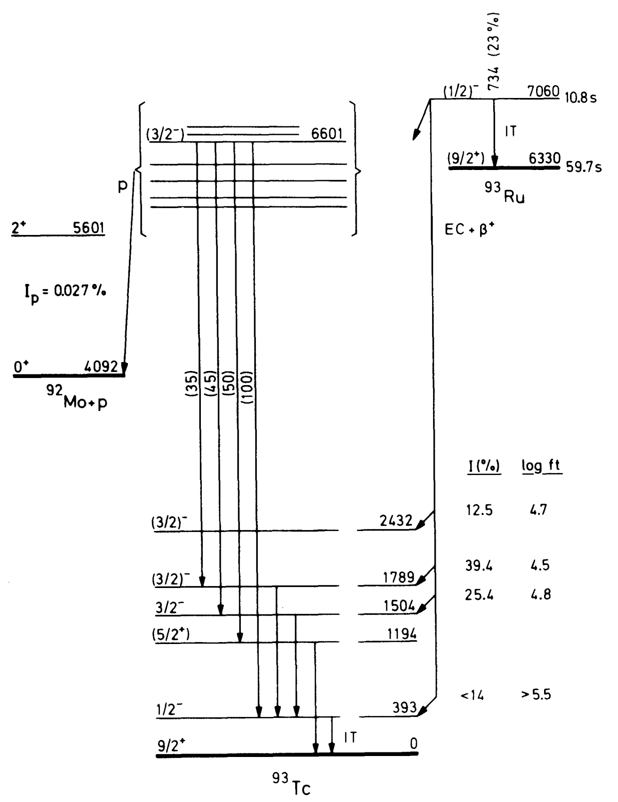

Fig. 4. A decay scheme for ^{93m}Ru. The β-decay branchings to the bound levels of ^{93}Tc are from the work of de Lange et al. [6]. On the left hand side the γ-decay pattern is shown for the resonance level at E_x = 6601 keV, which from the point of view of energy best matches the prominent delayed proton peak at 2481 keV.

4.2. Delayed proton spectrum

Due to the influence of the N = 50 shell, the first excited state ($J^\pi = 2^+$) of ^{92}Mo lies at 1.509 MeV. This greatly simplifies interpretation of the delayed proton spectrum of ^{93m}Ru, since only proton transitions leading to the ground state have to be considered. In the case of ^{99}Cd the situation is more complex, because the 2^+ state in ^{98}Pd is at 0.841 MeV and the energy window for proton emission is much wider in ^{99}Ag than in ^{93}Tc. For ^{73}Kr, in which energetic conditions are similar to those in ^{99}Cd, the fraction of delayed proton decays populating the first excited level has been measured to be 0.20 ± 0.04 [24].

As is evident from fig. 1(a), the statistical model calculation with a constant β-strength function fails to reproduce the observed delayed proton spectrum, although reasonable agreement is obtained for the total proton branching. Total widths of the proton emitting states are dominated by the proton decay channel for proton energies exceeding 2 MeV. The predicted alpha particle binding energy in ^{93}Tc is 5.535 MeV [23] and therefore no competition is expected from delayed alpha particle emission. Radiative widths for the levels under discussion are predicted to be of the order of 0.1-0.2 eV according to the formula given by Bartholomew et al. [25]. For delayed protons of 2.5 - 3.0 MeV the contribution of radiative widths to the total widths is then less than one tenth. Given the dominance of the proton decay channel, statistical fluctuations in the delayed proton spectra should only reflect fluctuations in β-transition probabilities. According to the Porter-Thomas law the normalized variance of the intensity fluctuations in the delayed proton spectrum should be 2 [15]. The high intensity of the 2481 keV peak indicates a larger intensity variation and suggests that structural factors connected to individual level properties are decisive in this case.

4.3. Beta decay of ^{93m}Ru

If the most pronounced peaks in the observed delayed proton spectrum are interpreted as arising essentially from single β-transitions to individual levels in the emitter nucleus, corresponding log ft values can be calculated and compared to log ft values for transitions to known bound levels in ^{93}Tc. In Table I we present just such a comparison of log ft values based on our particle spectroscopic data and on γ-spectroscopic data of de Lange et al. [6]. Because Γ_γ values for the proton emitting states have not been measured, the given log ft values should be taken as upper limits. All the log ft values in the table are suggestive of allowed β-transitions and the β-strengths associated with the prominent delayed proton groups are comparable to the strengths of transitions populating J = $1/2^-$ or $3/2^-$ bound states.

Table I. Experimental log ft-values of β-transitions to the $1/2^-$ and $3/2^-$ states in the N = 50 nucleus ^{93}Tc.

Level energy (keV)	J	log ft_{max}	Ref.
392.7	$1/2^-$	5.5 [a]	6)
1503.9	$3/2^-$	4.8 [a]	6)
1788.9	$3/2^-$	4.5 [a]	6)
2431.9	$3/2^-$	4.7 [a]	6)
6600±6	$(1/2^-)$,$3/2^-$	4.6	this work
6653	$1/2^-$,$3/2^-$	5.3	this work
6677	$1/2^-$,$3/2^-$	5.0	this work

[a] Corrected for the measured Q_{EC}-value

It would be of interest to compare the measured β-strengths with calculated strengths for configurations based on a shell-model approach including pairing correlations and a residual G-T interaction. Such a calculation has been done for β^+-emitters in a wide mass range including $J^\pi = 9/2^+$ ^{93}Ru, for which an enhancement in the β-strength was predicted at 7.5 MeV [26]. Unfortunately no similar calculation is available for the decay of ^{93m}Ru and a direct comparison with theory cannot be given on this point. From the experimental point of view there is a need for measurement of relative values of proton and γ-decay widths of resonant states in ^{93}Tc. With such information and with more detailed knowledge of the decay characteristics of relevant resonance levels, it may be possible to determine β-strengths of at least some individual levels at high excitation in ^{93}Tc, a unique case in the A∿100 mass region.

Acknowledgements

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