EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN PPE/90-59

7.5.1990

CM-P00052042

THE DECAY OF ^{31}Ar

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Keywords

RADIOACTIVITY $^{31}\text{Ar}(\beta^+, p), (\beta^+, 2p)$ [from $\text{Ca}(p, 3pxn)$ reaction]; measured β -delayed E_p , proton-proton coincidences. ^{31}Ar deduced decay channels. CaO target, surface barrier Si detectors. On-line mass separation.

Abstract

A search was made for two-proton emission from the ground state of ^{31}Ar . No events were observed and the upper limit for the branching ratio is 5%. Beta-delayed proton and two-proton emission from ^{31}Ar were observed from several intermediate states in ^{31}Cl .

IS-140

(Submitted to Nucl.Phys.A)

1. Introduction

Our knowledge about nuclei at the proton drip line has recently improved due to the use of intermediate energy heavy-ion fragmentation reactions at GANIL. The $T_z = -5/2$ series has been extended [1] and the decay of ^{31}Ar could be studied in detail [2]. Reviews of recent work on proton-rich light nuclei are given in ref. [3]. We have previously investigated the decay of the $T_z = -2, -3/2$ Ar-isotopes [4,5] and report here on a continuation of these studies to the decay of ^{31}Ar .

The challenge for a detailed study of this isotope was the possibility for decay via ground-state emission of two protons; a decay mode originally predicted by Goldansky [6] but not yet observed. No direct evidence for the existence of such a decay branch could be given in ref. [2] due to their relatively high background at low energies, where the two-proton events would occur, and to the presence of ^{29}S , the hypothetical 2p-daughter, as a contaminant in the radioactive ion beam. Arguments were given [2] against a strong branch based on the good agreement between the theoretical β -decay half-life and the experimental one (the separation energy for two protons is still uncertain since the Q_{EC} -value could not be measured).

We have searched for the two-proton decay of ^{31}Ar at the ISOLDE facility, which offers two experimental advantages. First, no ^{29}S is present as contamination from the target and ion source used; secondly, the radioactive beam is of low energy and can therefore be stopped in a thin foil. This allows the use of thin detectors in which the beta particles deposit less energy thus reducing the background at low energies.

2. The experiment

The nuclide ^{31}Ar was produced in a yield of 1 atom/s at the ISOLDE facility at CERN in spallation reactions with a $2.4\ \mu\text{A}$ beam of 600 MeV protons from the Synchro-cyclotron impinging on a calcium oxide target kept at 1300°C . The target construction has been described earlier [4].

The 60 keV Ar^+ beam from the ISOLDE on-line electromagnetic isotope separator was directed through the central hole of a $500\ \mu\text{m}$ annular Si detector and stopped in a $20\ \mu\text{g}/\text{cm}^2$ carbon foil. Two different detector set-ups were used. In the set-up depicted in fig. 1a (later referred to as set-up 1) a telescope, consisting of a $15\ \mu\text{m}$ and a $500\ \mu\text{m}$ Si detector, was placed opposite to the annular detector. In set-up 2 the ΔE detector of the telescope was removed, the beam was pulsed (fig. 1b) and both beam-on and beam-off spectra were recorded. The 15 ms delay before the start of data-taking in the second set of spectra enhanced the difference of the ^{31}Ar content in the two sets of spectra. In general half-lives shorter than 100 ms could be identified clearly by comparing the two data sets. The energy signals from the three detectors and the time difference between the two thick detectors were recorded in list-mode, in the first set-up also the absolute time of each event was stored. The energy calibration was done using the energies and intensities [5] of the beta-delayed protons from ^{33}Ar , produced with the same target.

3. Results

The data are presented as one-dimensional projections and scatter plots. Both are needed for a full understanding of the decay and are discussed in subsections 3.1 and 3.2. In subsection 3.3 details of the search for direct two-proton emission are given.

3.1 One-dimensional spectra

Fig. 2a shows spectra from the $15\ \mu\text{m}$ detector. The singles spectrum (dashed line) is above 2 MeV dominated by the completely or partially stopped alpha particles from long-lived Po isotopes, a contaminant present in the ISOLDE separator. This background is strongly suppressed when a coincidence with the annular detector and

an anti-coincidence with the E-detector of the telescope is required (full line). The activity below 2 MeV in the coincident spectrum stems from ^{31}Ar . At the lowest energies the spectrum consists of partially stopped protons, above which - clearest in the singles spectrum - the 1.4 MeV line observed previously [2] is seen (indicated with an asterisk). The intense broad bump just below 2 MeV will be shown in the next subsection to come from proton-proton coincidences. Note that its large coincidence efficiency, almost twice the solid angle subtended by the annular detector, supports this interpretation since coincidences are possible both with the second proton and the preceding beta particle. No positive evidence for beta-delayed alpha emission was obtained. An upper limit on the beta-delayed alpha branch from the IAS to the ground state of ^{27}P is given in table 1.

Fig. 2b shows the spectrum from the telescope (set-up 1) in coincidence with low-energy events in the annular detector. The energies and relative intensities of the main peaks are given in table 1. The table also includes the proton energies from ref. 2, transformed to the laboratory system. The two values for the energy of the strongest proton line do not agree. As mentioned, our set-up was calibrated with single proton lines from ^{33}Ar ; these include two well-determined lines [5] at 1634 keV and 3167 keV, so our energies should be accurate. In ref. 2 the detected energy is the sum of contributions from the proton, the recoiling ion and the beta particle. The paper does not give details on the extraction of the transition energies (see ref. 7 for a short discussion of the pulse height defect associated with the recoiling ion). We do not see any narrow lines at 6000 keV, but a broad unresolved line here would still be consistent with the observation in ref. 2. Our relative intensities have been taken from the singles spectra wherever possible. These are somewhat marred by background coming from Po and by radioactive O and N isotopes in molecules of mass 31. To reduce the influence of the contaminants the singles spectra recorded during beam-off with set-up 2 were used for background subtraction. The remaining activity above 2.5 MeV in fig. 2b is also attributed to ^{31}Ar as both the background and the number of random coincidences are negligible here.

Part of this unresolved intensity stems from beta-delayed two-proton decays where only one of the emitted protons is detected in the telescope. Two protons hitting the telescope simultaneously will have a different relative energy loss in the ΔE detector than a single proton. The two types of events are therefore clearly separated in a ΔE -E plot. The background in such a plot was seen from a run with ^{33}Ar to be negligible so that events with two-proton summing in the telescope could be easily extracted (fig. 2c). The peaks are broadened due to the energy distribution in the (not detected) recoil nucleus and due to summing with the preceding beta particle. Only the peak at 7.16 MeV, also visible in fig. 2b, is clearly resolved and must originate in the two-proton decay from the isobaric analogue state (IAS) in ^{31}Cl .

3.2 Two-dimensional spectra

The scatter plots in fig. 3 were obtained with set-up 1 and have the energy recorded in the annular detector as abscissa. The ordinate in the main plot is the summed energy in the telescope, in the inset it is the energy of particles stopped in the thin detector. In the off-line analysis events coming from beta particles in the telescope were suppressed; this gives the cut-off at 1 MeV in the main figure. Particle-beta and beta-beta coincidences are seen along and close to the axes; true particle-particle coincidences are spread over the entire plot. Strongest among the latter are the two clusters in the inset appearing with the high energy particle just below 2 MeV. The low energy particle in these clusters is clearly separated from the betas. The clusters must therefore be due to beta-delayed sequential two-proton emission (the high-energy proton is not stopped completely in the 15 μm detector, thus distorting the response slightly). The energies of the two proton lines are 445 and 1780 keV as obtained from the annular detector whose calibration was the most accurate. The 1780 keV line has a width of 220 ± 80 keV and thus corresponds to the intense broad bump seen in fig. 2a.

No other clusters are seen. There are, however, many coincidences almost evenly distributed with a total summed energy from 2.5 MeV up to 8 MeV. These are distributed in the same way in the telescope as in the annular detector and are

thus true proton-proton coincidences, as they are identified as protons in the telescope. Even very narrow $\beta 2p$ -transitions will have a dispersion in the measured energy of several hundred keV for the following reasons: The detectors cover about 2π sr which means that the recoil nucleus as well as the beta particle will be detected in some events. Furthermore, the summed energy of the two protons will vary due to the three-body final state. One would therefore expect the coincidences to fall in "bands" of approximately constant sum-energy. With the present statistics only the highest of these bands may be identified. This band, which in fig. 3 is indicated with a hatched area as a guide for the eye, must be due to two-proton emission from the IAS to the ground state of ^{29}P . Lower lying coincidences must also be due to $2p$ branches: to excited states in ^{29}P , from lower states in ^{31}Cl or both. Several branches are present, but the statistics is too low to separate them.

Note that the high energy $2p$ -events rather form bands than clusters as the $2p$ events at 485 keV and 1780 keV. The decay from the high lying states in ^{31}Cl is therefore most likely non-sequential, see ref. [3] for a discussion of possible decay mechanisms. The $\beta 3p$ branch reported in ref. [7] could contribute below a total sum-energy around 5 MeV and therefore makes the interpretation of our events in this region more ambiguous. The excitation energy of the IAS in ^{31}Cl can be found from the energies of the events in the highest band (with due corrections for beta-summing and for the energy of the recoil nucleus). The cm-energy for this $2p$ -decay can also be found from the peak near 7 MeV in fig. 2b and 2c. These estimates coincide within the errors and give $E_{\text{cm}}(2p) = 7.7 \pm 0.1$ MeV, which corresponds to $E_{\text{IAS}}(^{31}\text{Cl}) = 12.39 \pm 0.12$ MeV. Independent evidence for the $2p$ decay of the IAS was obtained in recent experiments at GANIL [7] and was also reported in ref. [8].

As the angular distribution of the two protons in the $\beta 2p$ decays has not been measured, we cannot deduce the branching ratios for these decays (except for the low-energy proton-proton coincidences, where the 1780 keV line is seen in the singles spectra). Only uncorrelated two-protons will be seen in fig. 3 due to the 180° geometry. The spectrum in fig. 2c could also include contributions from diproton (^2He) emission, if this branch exists. It would lead to strongly correlated protons, for the IAS branch, e.g., one has a maximum opening angle of 30° between the protons. Correcting for the solid angle we do see 50% more two-protons from the IAS in the 0° geometry than in the 180° geometry, but this is only a one standard deviation result and therefore not conclusive.

3.3 Two-proton radioactivity

The beta-delayed proton decay of ^{29}S , the two-proton daughter of ^{31}Ar , can be used in two different ways to search for $2p$ radioactivity from the ^{31}Ar ground state. The energies and branching ratios in the ^{29}S decay were taken from the work of Vieira et al. [9].

In the first approach, we compare the intensity at the position expected for the most intense branch of ^{29}S (5.44 MeV, marked in fig. 2b with an arrow) with the intensity of the strongest proton line from the decay of ^{31}Ar at 2.08 MeV. No counts were observed around 5.44 MeV, while the 2.08 MeV line contained 152 counts. The coincidence efficiency is larger at 5.44 MeV than at 2.08 MeV, but we neglect this to be conservative. The 95% confidence upper limit on the number of counts in the ^{29}S line (a 15.8% branch) is 2.3, this yields a ratio of branching ratios for ^{31}Ar of $R = b(2p)/b(2.08 \text{ MeV}) = 0.096$.

We also tried to search directly for the two-proton decay with set-up 1. The signal in the ΔE detector (where a possible $2p$ event would be detected) was used as a start signal to look for protons from ^{29}S in the other detectors. No such events were seen within two half-lives of ^{29}S , i.e. 376 ms. Corrections were made for the decay of ^{29}S outside the time window, the proton branching ratio of ^{29}S and the detection efficiency for $2p$ -events. Normalization to the number of protons in the 2.08 MeV line (so that the efficiency for proton detection drops out) gives a 95% confidence upper limit on R of 0.28. This limit only applies for a summed energy of the two protons larger than 300 keV as this energy was chosen as cut-off in the ΔE detector. The detection-efficiency for the two-protons was taken to be 0.1, the square of the solid angle of the ΔE detector. This would be appropriate

for uncorrelated two-proton decays and is an underestimate if diproton emission occurs (see e.g. the discussion of correlations in ref. [10]).

With a branching ratio $b(2.08 \text{ MeV})$ around 30% [2] our upper limit on R constrains $b(2p)$ to be less than 3%. From the total observed intensity, summing all the entries in table 1, we obtain our adopted experimental limit of 4.5% (note that the expected branch to the ground state of ^{31}Cl is unobserved and thus not included in this estimate).

4. Discussion

The major outcome of our work is the limit on the branching ratio for direct two-proton emission relative to the strongest line from ^{31}Ar , $b(2p)/b(2.08 \text{ MeV})$, of 9.6%. In addition several beta-delayed proton and two-proton branches were observed. It is, however, not in all cases possible to deduce the corresponding levels in the decay scheme. From the schematic decay scheme in fig. 4 it is evident that many excited states in the one- and two-proton daughters must be fed. The identification of the proton-emitting levels would in general require e.g. pp coincidence data. The cases, where identification of the proton emitting states is possible, are treated in the following.

Consider first the proton-proton coincident events from the inset of fig. 3. The two-proton decay is here sequential and the inset in fig. 4 shows three possible decay routes. Route I can be discarded, as the low energy proton has a very low penetrability factor - if emitted first it would be dwarfed by the much more probable transitions to lower states in ^{30}S . Analogous arguments show that the state fed by the low energy proton must be the ground state of ^{29}P . If not, the decay would be a competition between a low and a high energy proton (shown as decay route III in the figure) and the high energy proton would strongly dominate even when unfavoured by nuclear structure. The $\beta 2p$ decay must therefore take place via two new levels: a 220 keV wide level at 6.92 MeV in ^{31}Cl and a narrow level around 4.85 MeV in ^{30}S . We shall look at the two levels in turn to verify the plausibility of this assignment.

The ground state of ^{29}P has $I^\pi = 1/2^+$. From a comparison with the mirror nucleus ^{30}Si one would expect that the 4.85 MeV level in ^{30}S has $I^\pi = 2^+, 3^+$. The low energy proton is therefore probably emitted as a d-wave. An s-wave cannot be excluded, but is less likely. The partial half-life for proton decay is estimated from the formulas [11]

$$\Gamma = \frac{2P_1 \gamma^2}{1 + \gamma^2 \frac{1}{dE}} , \quad \gamma^2 = \frac{\hbar^2}{Ma^2} \theta^2 \quad (1)$$

where P and S are the penetration and shift factors for angular momentum l , M the reduced mass, a the channel radius and θ^2 the dimensionless reduced width. For θ^2 around 1 the partial half-life is 5-10 fs for a d-wave and 0.1-0.2 fs for an s-wave. Competition from gamma decay of the 4.85 MeV level can be estimated from the partial half-lives for gamma decay for close lying levels [12] that are around 100 fs. This is consistent with the experimental observation that proton emission dominates for the 4.85 MeV level, although the dimensionless reduced width must be rather high if the proton is in a d-wave.

The 6.92 MeV level in ^{31}Cl is fairly broad. If the only decay channel would lead to the 4.85 MeV level in ^{30}S the dimensionless reduced width θ^2 from eq.(1) should again be close to 1. One could, however, also expect strong proton branches to lower levels in ^{30}S . They would give broad singles proton peaks at 6.4, 4.3, 3.1 and 2.9 MeV. Fig. 2b has unassigned counts in these energy regions, which could originate from such transitions. The rest of the intensity is due to the $\beta 2p$ decays mentioned earlier or to unresolved weaker lines.

The large relative intensity for the observed decay route shows that ^{31}Ar has a high beta branching ratio to the 6.92 MeV level. The strong $\beta 2p$ branch is compatible with previously published data: In the set-up used in ref.[2] the two

low-energy protons are summed, they thus contribute to the broad peak between 2 and 2.5 MeV in their fig. 3. It should be pointed out that similar intense broad peaks were seen (at a slightly lower excitation energy) in the decay of ^{33}Ar [5].

From the measured energy of the IAS we can estimate the Q-value of ^{31}Ar . The Coulomb energy difference, ΔE_C , between ^{31}Cl and ^{31}Ar is obtained from an extrapolation of the experimental [13] Coulomb energy shifts between $^{32,33}\text{Cl}$ and $^{32,33}\text{Ar}$ using the trend given in eq.(5) of ref. [14]. This gives $\Delta E_C = 7.01 \pm 0.10$ MeV. The Q-value of ^{31}Ar becomes 18.62 ± 0.16 MeV, corresponding to a two-proton separation energy of $S_{2p} = -0.13 \pm 0.18$ MeV and a mass excess of 11.56 ± 0.17 MeV.

An independent limit on the Q-value is obtained from the limit on the direct 2p branch. This corresponds to a partial half-life for this decay mode larger than 0.25 s; extrapolating from the calculations of ref.[10] this implies that $S_{2p} > -0.4$ MeV, which gives $Q < 19.0$ MeV. The two estimates are consistent.

In summary, the new features found in this study of the decay of ^{31}Ar are the strong beta-delayed two-proton branches, occurring many MeV below the isobaric analogue state, and the suggested non-sequential decay mechanism for the most energetic of these branches. We have no indication of a diproton decay mechanism. Fig. 4 shows the deduced partial decay scheme and table 1 gives all the resolved lines and our limits on unobserved decay modes.

Acknowledgements: We are grateful to O. Jonsson and K. Schlösser for help with the experiment, and to D. Bazin, V. Borrel, P.G. Hansen and A.C. Mueller for discussions.

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Table 1.
Observed lines^a from ³¹Ar.

	E _{lab} , keV		Relative intensity	Branch
	this work	ref.2		
Narrow lines	1425±15	1471± 39	24 ±7	
	2080± 8	2119± 10	≡ 100	
	3636±10	3580±100	6.6±1.5	
	4010±10		8.8±1.5	
	5240±20	5275±100	10 ±3	
		6000±100		
	7160±20		d	IAS → 2p
Limits ^b			< 1.5	IAS → α
			< 9.6	direct 2p
Proton-proton coinc.	445±15 1780±40	}	50 ± 15	
	c		8±3 ^e	IAS → 2p

^a The unresolved structures are not included, their total relative intensity is at least 40.

^b No single proton lines from the IAS were identified. Those to the ³⁰S g.s. would have an energy of 11.7 MeV, in the present experiment no detector of sufficient thickness to stop such protons were used.

^c Continuous distribution, E_{c.m.}(2p) = 7.7±0.1 MeV.

^d See bottom line in table.

^e Assuming an isotropic distribution of the two protons.

Figure captions

- Fig. 1. Schematic view of the two set-ups. A 500 μm thick annular Si detector was placed opposite to (a) a telescope consisting of a 15 μm and a 500 μm Si detector or to (b) a 500 μm detector. In set-up 2 the beam was pulsed according to the scheme shown in the lower part of figure b.
- Fig. 2. Spectra taken with set-up 1 (fig. 1a).
 (a) Spectrum from the 15 μm detector. The dashed line shows the singles spectrum, the full line shows the spectrum in coincidence with signals from the annular detector and in anti-coincidence with the 500 μm E-detector. The 1.4 MeV line is marked with an asterisk, the broad bump above it is due to 2p events.
 (b) Spectrum from the telescope in coincidence with low-energy events in the annular detector. The position of the 2p-line from the IAS and the expected position for the strongest proton line from ^{29}S are indicated.
 (c) The two-proton sum spectrum in the telescope with events identified from the E- ΔE distribution.
- Fig. 3. Scatter plot of coincident events recorded during a total measuring time of 18.6 hours. Only events with a proton in the telescope are shown, the band along the ordinate consists therefore of beta-proton coincidences whereas the plane contains proton-proton coincidences with a summed energy of the two particles up to 8 MeV. The hatched area contains events from decays through the IAS. In the inset the events that gave a signal only in the thin front detector are shown. The bands along the axes contain beta-beta and beta-proton coincidences, the events in the upper right corner stem from contamination alphas that go through the thin detector.
- Fig. 4. Decay scheme of ^{31}Ar . The deduced decay channels and levels are shown in full lines, possible channels are indicated with dashed lines. The inset shows possible decay routes of the low-energy two-proton decay where the decay route II is the most plausible one (see text).

Fig. 1

Fig. 2

Fig. 3

Fig. 4