

Alpha-decay of neutron-deficient ^{200}Fr and heavier neighbours

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Abstract. A new measurement of the ^{200}Fr α -decay half-life, with improved accuracy compared to previous measurements, has been carried out at the Isolde-CERN on-line mass separator. A half-life of 49(4) ms has been obtained, which is substantially different from earlier literature values. For the ^{196}At daughter decay, a half-life of 389(54) ms and an α branching ratio $b_\alpha = 94(5)\%$ were measured. In addition, the half-lives of $^{201,203,205}\text{Fr}$ and $^{197,199}\text{At}$ are reported.

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1 Introduction

In the neutron-deficient nuclei around $Z = 82$, shape coexistence phenomena induced by proton excitations across the shell gap have been observed [1,2]. A large amount of experimental data has been obtained through α - and β -decay studies as well as in-beam studies, focussing mainly on even-even nuclei. In the odd and odd-odd mass nuclei, proton excitations across the $Z = 82$ shell gap give rise to isomerism, which can be investigated via α -decay in *e.g.* the francium isotopes which disintegrate through the corresponding astatine and bismuth to thallium nuclei (see, *e.g.*, [3]). A renewed interest in the study of very neutron-deficient francium isotopes resulted in conflicting results with respect to reported half-lives and to the observation of isomeric α -decay. In particular, the α -decay of ^{200}Fr was investigated in fusion-evaporation reactions at the recoil separators GARIS (RIKEN) [4] and RITU (JYFL) [5]. Both experiments report the observation of ^{200}Fr α -decay, but with very different half-life values: $E_\alpha = 7500(30)$ keV and $T_{1/2} = 570_{-140}^{+270}$ ms in [4] and $E_\alpha = 7468(9)$ keV and $T_{1/2} = 19_{-6}^{+13}$ ms in [5]. These results were

obtained by requiring spatial and temporal correlations of the implantation of the mother nucleus, its α -decay and that of the daughter nucleus. Only six correlated chains were observed in each experiment. In this paper we report on a new study of the very neutron-deficient francium and astatine isotopes using intense high-quality sources produced with an on-line mass separator.

2 Experimental details

The experiment carried out at the ISOLDE on-line mass separator [6], used a 1.4 GeV pulsed proton beam (2.9×10^{13} protons/pulse) impinging on a 51 g/cm² ThC_x/graphite target. After diffusion out of the 2150 °C hot target, the reaction products were guided to a 2050 °C hot niobium cavity. Due to their low ionization potential (4.07 eV), the francium atoms were efficiently ionized by surface ionization in the hot cavity. After mass separation by means of the General Purpose Separator [6], the 60 keV $A = 200$ beam was implanted into a 20 $\mu\text{g}/\text{cm}^2$ carbon foil. After proton impact on the target, ions were implanted for 80 ms and the subsequent decay was recorded for 1120 ms, which was the time left before the next proton pulse.

Data were also collected at masses 201, 203 and 205. For these measurements the time sequence was altered: the implantation/decay cycles were 100 ms/1100 ms,

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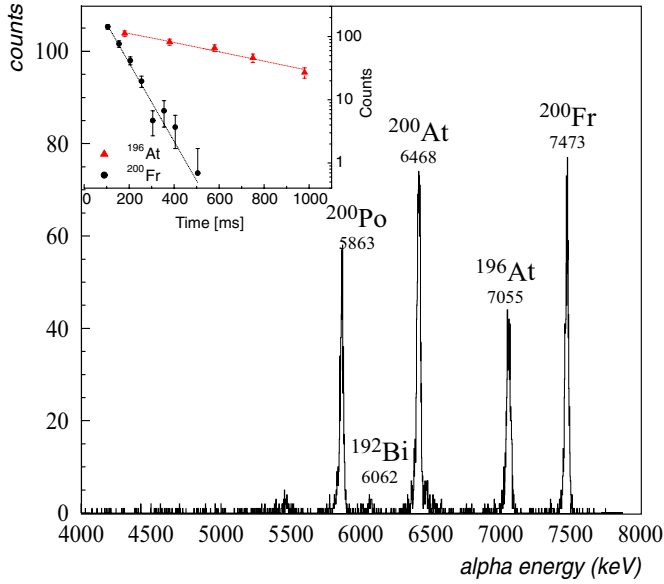


Fig. 1. The α spectrum recorded at $A = 200$. The energy labels are in keV. The inset shows the time behaviour for the ^{200}Fr and ^{196}At activities.

1000 ms/ 15800 ms and 40 ms/16760 ms for $A = 201$, 203 and 205, respectively.

Behind the carbon foil, a Si detector (area 150 mm², thickness 300 μm for $A = 200, 203, 205$; area 450 mm², thickness 1000 μm for $A = 201$) was placed at a distance of 7 mm. For calibration of the Si detector the following isotopes were used: ^{196}At ($E_\alpha = 7055(7)$ keV [7]), ^{200}Po ($E_\alpha = 5863(2)$ keV [8]), ^{203}Fr ($E_\alpha = 7133(4)$ keV [7]), ^{197g}At ($E_\alpha = 6960(5)$ keV [9]), ^{197m}At ($E_\alpha = 6707(5)$ keV [9]) and ^{199}At ($E_\alpha = 6643(3)$ keV [7]). These isotopes were produced in irradiations or as daughter activities at different masses. The α energy resolution was 30 keV FWHM for the data collected at mass 200, 203, 205 and 55 keV in the case of $A = 201$.

3 Experimental results and discussion

3.1 ^{200}Fr decay chain

Figure 1 shows the α energy spectrum obtained at mass 200 in ~ 3 hours of data collection. The α lines at 5863 keV and 6468 keV are attributed to ^{200}Po and ^{200}At , respectively. Despite a larger N/Z ratio, leading to a three to four orders of magnitude higher production rate in the target, as compared to ^{200}Fr , the ionization efficiency for these isobars is substantially lower due to their much higher ionization potential (8.42 and 9.65 eV, respectively). The measured α -decay energy of ^{200}Fr is 7473(12) keV, in agreement with the literature values (see table 1). The yield for ^{200}Fr was 2×10^{-1} ions/s in our experiment, compared to 3×10^{-4} ions/s in [5].

The inset in fig. 1 shows the time behaviour of the α intensities for the ^{200}Fr and ^{196}At lines. The data for

Table 1. Energies and half-lives for α lines accompanying the Fr and At decays.

Isotope	Energy (keV)	$T_{1/2}$	Reference
^{200}Fr	7473(12)	49(4) ms	this work
	7500(30)	570_{-140}^{+270} ms	[4]
	7468(9)	19_{-6}^{+13} ms	[5]
^{196}At	7055(12) ^(a)	389(54) ms	this work
	7053(30)	320_{-90}^{+220} ms	[4]
	7044(7)	390_{-120}^{+270} ms	[5]
	7048(5)	388(7) ms	[10]
	7065(30)	253(9) ms	[11]
	7055(7)	300(100) ms	[12]
^{201g}Fr	7379(7)	67(3) ms	this work
	7361(7)	69_{-11}^{+16} ms	[5]
	7369(8)	53(4) ms	[13]
	7388(15)	48(15) ms	[14]
^{201m}Fr	7454(8)	19_{-6}^{+19} ms	[13]
^{197g}At	6963(4) ^(a)	390(16) ms	this work
	6956(5)	370_{-60}^{+90} ms	[5]
	6960(5)	388(6) ms	[9]
	6959(6)	340(20) ms	[13]
	6957	350(40) ms	[15]
	^{197m}At	6707(5)	2.0(2) s
^{203}Fr	7132(5) ^(a)	0.560(15) s	this work
	7133(4)	0.55(2) s	[7]
^{199}At	6643(4) ^(a)	6.92(13) s	this work
	6643(3)	7.2(5) s	[7]
^{205}Fr	6916(5)	3.80(3) s	this work
	6915(4)	3.85(10) s	[7]
^{201}At	6343(4)		this work
	6344(2)	89(3) s	[7]

^(a) This α line has been used for energy calibration.

^{200}Fr were fitted using a single exponential function combined with a constant background. The background, deduced from the part of the spectrum between 430 ms and 1200 ms after proton impact, represents 2.5% of the total intensity of the 7473 keV line. A half-life value of 49(4) ms is obtained, at variance with both the value obtained by Enqvist *et al.* [5] and the earlier value by Morita *et al.* [4]. The time behaviour of the ^{196}At α line intensity was fitted with a mother-daughter decay function; a half-life of 389(54) ms was deduced, which agrees with the earlier measurements, [4, 5] and [10], but differs significantly from the value of Pu *et al.* [11], which used a fusion-evaporation reaction combined with the genetic correlation technique, similar to [4] and [5]. All data are summarized in table 1.

The α branching ratio for ^{196}At (only present as the daughter of ^{200}Fr) is obtained by comparing mother and daughter α intensities. After α -decay, the daughter nuclei might recoil out of the catcher foil, decreasing the observed daughter activity. It should be noted that the recoil of daughter activity into the detector, enhancing the α detection efficiency for daughter activity, does not

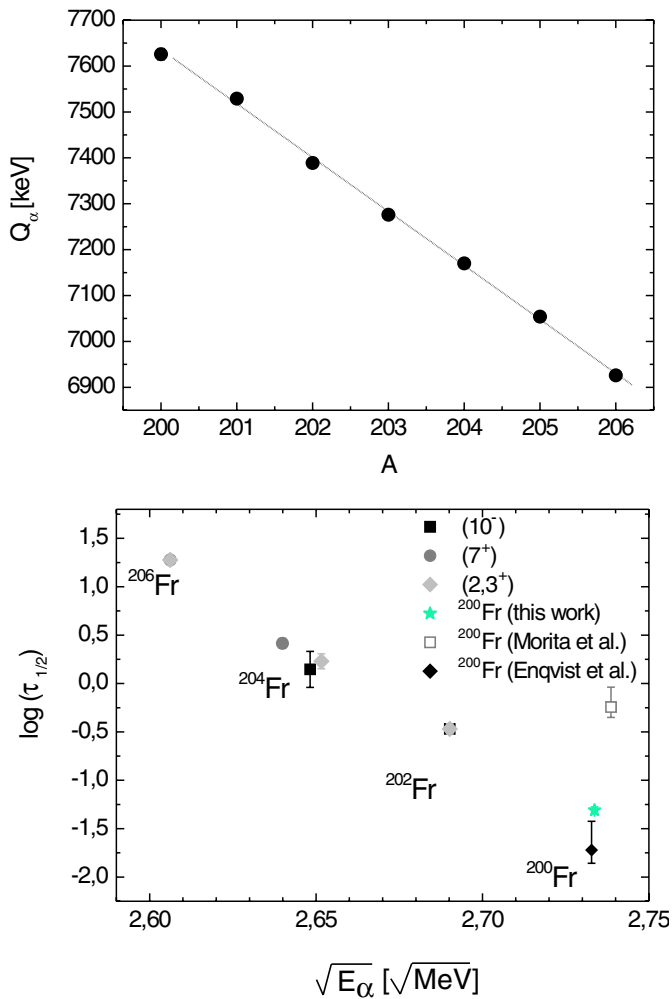


Fig. 2. Top: Q_α -systematics for the francium isotopes $^{200-206}\text{Fr}$. The straight line is shown to guide the eye; Bottom: Geiger-Nuttall plot for the even- A isotopes, $^{200-206}\text{Fr}$.

have to be considered as the thickness of the catcher foils is $20 \mu\text{g}/\text{cm}^2$, compared to an average implantation depth of $8 \mu\text{g}/\text{cm}^2$. A 25% recoil loss correction [16], calculated with the SRIM simulation code [17], was made. The final calculation yields an α branching ratio of 94(5)% for the decay of ^{196}At .

The α -decay of ^{196}At proceeds to ^{192}Bi . The α lines from the decay of the 10^- ($E_\alpha = 6052$ keV) and $(2, 3)^+$ ($E_\alpha = 6060$ keV) isomers of ^{192}Bi [18] could not be resolved within the experimental energy resolution. A spin assignment based on the α branching ratios of 12(5)% for the low-spin and 10(3)% for the high-spin isomer of ^{192}Bi [18] was also not feasible.

In the heavier odd-odd francium isotopes ($A = 202, 204, 206$), two or three isomeric, α -decaying states were identified [3]. In ^{200}Fr only one α branch could be identified in this study. The α energy is the same in all measurements ([4,5] and this work), within the experimental accuracy. However, one cannot exclude a doublet structure of this α line, like, *e.g.*, in ^{202}Fr [3]. In ^{202}Fr , both known α -decays have similar decay properties ($E_\alpha = 7237$ keV

and $T_{1/2} = 0.34$ s) and feed two α -decaying isomers in ^{198}At , with different half-lives and tentative spin assignments of 3^+ and 10^- . A similar structure in ^{200}Fr cannot be excluded, but the observation of only one α branch in the daughter makes this assumption unlikely.

Three different half-life values have been measured for the ^{200}Fr decay. However, the result obtained at RITU [5] and our data follow the systematics of the heavier isotopes, shown in a Geiger-Nuttall plot in fig. 2. The value of [5] has been determined on the basis of 6 events and meets the problem of estimating the accidental correlations which could affect the half-life determination. In our experiment, the good quality of mass-separated ISOL beams was exploited and the statistics were about 50 times higher.

The production mechanism used in the present work, high-energy proton-induced spallation of ^{232}Th , is different from the heavy-ion fusion-evaporation reactions used in [4], $^{169}\text{Tm}(^{36}\text{Ar}, 5n)$ and [5], $^{170}\text{Yb}(^{35}\text{Cl}, 5n)$, which might lead to the population of different isomers in this study compared to [4] and [5]. However, a very different isomer population in the similar production mechanisms of [4] and [5] is not expected. In conclusion, our data do not show any evidence for α -decay from an isomeric state in ^{200}Fr .

3.2 Odd- A francium isotopes: $^{201,203,205}\text{Fr}$

Additionally, data were taken on the odd- A francium isotopes $^{201,203,205}\text{Fr}$. The obtained energies and half-lives are given in table 1. In these nuclei, as in *e.g.* odd- A neutron-deficient bismuth and astatine isotopes, a $1/2^+$ shell model intruder state, resulting from a proton excitation from the $\pi s_{1/2}$ orbital, is expected and was observed for the first time at the RITU separator [13] in ^{201}Fr . A compilation of the presently available experimental data is shown in fig. 3, presenting the decay scheme for ^{201}Fr .

The energy spectrum obtained at mass 201 is shown in fig. 4. Apart from the known α lines at 6325 keV from ^{201}At , 7379 keV from ^{201g}Fr and 6963 keV from ^{197g}At , the α transition of 6707 keV from the $1/2^+$ intruder state from ^{197m}At [15] was observed. The presence of the latter α line at mass 201 can be explained by feeding from the α -decay of the ^{201}Fr isomer as observed in [13]. Indeed, the small shoulder at the high-energy side of the 7379 keV α line is probably caused by the presence of the 7454 keV α line of ^{201m}Fr . The extra α counts observed around 6475 keV could stem from the known α line of the decay of ^{193m}Bi and are consistent with the decay from the $1/2^+$ intruder states. Feeding from the ^{201}Fr ground state to the $1/2^+$ state in ^{197}At cannot be excluded but does not account for the observed intensity (see below).

The isomer ratio obtained for the production of the low-spin state relative to the high-spin state in ^{197}At was 3% (assuming $b_\alpha = 100\%$ for ^{197m}At). This value is comparable to the ratios obtained in heavy-ion reactions: $< 1\%$ in [15] and $3.9 \pm 1.2\%$ in [9].

A careful investigation of the time behaviour of the ^{201g}Fr and ^{197g}At α lines gave half-life values of 67(3) ms and 390(16) ms, respectively. The data are summarized

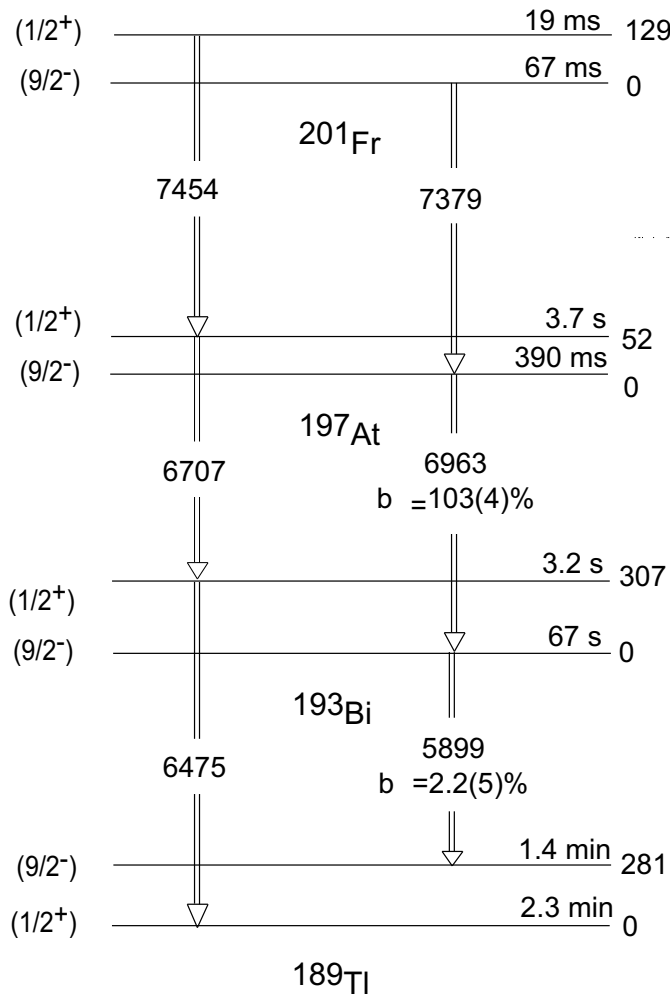


Fig. 3. Decay scheme of ^{201}Fr and ^{197}At . The α -decay energy as well as the half-life for $^{201\text{m}}\text{Fr}$ are taken from [13]. E_α and $T_{1/2}$ for $^{201\text{g}}\text{Fr}$ and $^{197\text{g}}\text{At}$ are taken from this work. The excitation energy of the $(1/2^+)$ state in ^{197}At has been taken from [15], its half-life from [9]. The $^{197\text{g}}\text{At}$ and $^{193\text{g}}\text{Bi}$ α branching ratios have been determined in this work. Other data are from [8].

in table 1. The α transition at 5882 keV was attributed to the decay of the $^{193\text{g}}\text{Bi}$ ($9/2^-$) ground state [7]. From the intensity ratio of the $^{201\text{g}}\text{Fr}$, $^{197\text{g}}\text{At}$ and $^{193\text{g}}\text{Bi}$ α lines, and taking into account the recoil loss correction [16], an α branching ratio of 103(4)% for $^{197\text{g}}\text{At}$ and 2.2(5)% for $^{193\text{g}}\text{Bi}$ was obtained, consistent with the limits given in [15]. The corresponding reduced widths (assuming $\Delta L = 0$ transitions) are 64(4) keV, 51(3) keV and 18(4) keV and the deduced hindrance factors, comparing with the even-even neighbours, are 1.3(2), 2.1(3) and 2.7(6) for $^{201\text{g}}\text{Fr}$, $^{197\text{g}}\text{At}$ and $^{193\text{g}}\text{Bi}$, respectively, indicating that these α transitions are unhindered and link states with the same configuration, spin and parity. For $^{201\text{g}}\text{Fr}$, the hindrance factor is determined relative to ^{200}Rn only, as the reduced width for its second even-even neighbour, ^{202}Ra is only poorly known. In addition, upper limits for the branching ratios of the $9/2^- - 1/2^+$ fine-structure transitions have been calculated to be 2% for the $^{201\text{g}}\text{Fr}$ decay

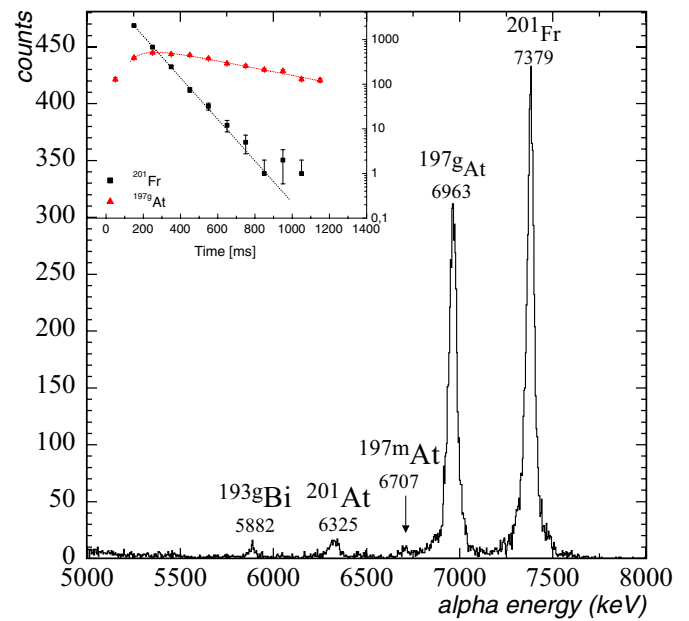


Fig. 4. Energy spectrum taken at $A = 201$. The energy labels are in keV. The inset shows the time behaviour for the $^{201\text{g}}\text{Fr}$ and $^{197\text{g}}\text{At}$ activities.

and 0.5% for the $^{197\text{g}}\text{At}$ decay. The corresponding lower limits of the hindrance factors (assuming $\Delta L = 0$ transitions) are 30 and 13, respectively, relative to the $9/2^- - 9/2^-$ transitions. These numbers are consistent with experimental data on similar transitions in the α -decay of bismuth isotopes [19], where hindrance factors an order of magnitude larger than the lower limits are reported.

4 Conclusion

In this paper the α -decay of neutron-deficient francium and astatine isotopes was studied. Half-lives were remeasured for $^{200,201,203,205}\text{Fr}$ and $^{196,197,199}\text{At}$. For ^{200}Fr , the half-life result obtained in this work differs substantially from previous studies and no evidence for isomeric α -decay was found.

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