## Isomers in three doubly odd Fr-At-Bi $\alpha$ -decay chains

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The  ${}^{206}\text{Fr} \rightarrow {}^{202}\text{At} \rightarrow {}^{198}\text{Bi}$ ,  ${}^{204}\text{Fr} \rightarrow {}^{200}\text{At} \rightarrow {}^{196}\text{Bi}$ , and  ${}^{202}\text{Fr} \rightarrow {}^{198}\text{At} \rightarrow {}^{194}\text{Bi}$  a-decay chains have been studied by standard spectroscopic techniques using an on-line isotope separator. All the studied doubly odd isotopes have at least two isomers, which decay by a combination of the following decay modes: a emission,  $\beta^+/\text{EC}$  (electron capture) decay, and internal transition (IT). The internal transition, a highly retarded E3, is the *j*-forbidden transition between the  $[\pi h_{9/2} \otimes v i_{13/2}]_{10^-}$  and the  $[\pi h_{9/2} \otimes v f_{5/2}]_{7^+}$  states. The B(E3) values of these IT's together with their energy behavior as a function of the neutron and proton number, compared to the energy difference between the  $\frac{13}{2}^+(v i_{13/2})$  and  $\frac{5}{2}^-(v f_{5/2})$  states in the odd-mass Pb isotones, indicate that these proton-neutron-coupled states have a rather pure shell-model character.

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#### I. INTRODUCTION

The neutron-deficient nuclei around the Z=82 shell closure show at low and medium energy a rich variety of different excitations. One of the well-known phenomena is the occurrence of shape coexistence, related to shellmodel intruder states [1,2]. The region below lead is studied in great detail by using different kind of methods such as in-beam techniques,  $\alpha$  and  $\beta^+$ /EC (electron capture) decay studies, laser spectroscopy, etc. Above the shell closure, the situation is much more difficult. Inbeam spectroscopy is very much hindered by the fission and charged-particle evaporation channels. For instance, in the polonium isotopes the lightest system studied inbeam is <sup>196</sup>Po [3]. Information from  $\beta^+$ /EC-decay studies is also rather scarce as there is strong competition from  $\alpha$  decay. Most of the isotopes in this neighborhood have been identified by their  $\alpha$  decay but fine structure in the  $\alpha$  decay has only been intensively studied in some special cases, for instance, to identify  $0^+$  intruder states in even-even lead [4] and polonium [5] isotopes.

We have studied at the LISOL isotope separator three  $\alpha$ -decay chains: from <sup>206,204,202</sup>Fr over <sup>202,200,198</sup>At to <sup>198,196,194</sup>Bi. Several new isomers have been found and in most of the cases the relative position of the isomers has been determined on the basis of the interconnecting internal transition (IT). The  $\alpha$ -branching ratios of the different isomers have been obtained by comparing mother to daughter  $\alpha$ -decay intensities of chemically pure, mass-separated sources or by comparing the  $\alpha$  activity to the other decay modes such as IT and  $\beta^+$ /EC decay. The  $\beta^+$ /EC decay of <sup>202,200</sup>At to <sup>202,200</sup>Po has also been studied and a level scheme for <sup>202,200</sup>Po has been constructed.

#### **II. EXPERIMENTAL DETAILS**

The francium and astatine isotopes were produced in a heavy-ion fusion reaction and mass-separated with the Leuven Isotope Separator On-Line (LISOL) facility [6]. The <sup>202,204,206</sup>Fr isotopes were produced in the

<sup>nat</sup>Ir(5 mg/cm<sup>2</sup>)[<sup>20</sup>Ne(
$$< 245 \text{ MeV}$$
), xn]

and

$$^{181}$$
Ta(8 mg/cm<sup>2</sup>)[ $^{32}$ S( < 280 MeV), 2pxn ]

reactions while the

$$^{nat}$$
Re(16 mg/cm<sup>2</sup>)[<sup>20</sup>Ne( < 245 MeV), xn]

reaction was used to produce <sup>198,200,202</sup>At. In order to obtain an optimum production rate for the different nuclei the beam energy was degraded by putting tantalum degrader foils (2.5  $\mu$ m) in the beam just in front of the target. The Febiad ion source was used in two modes. In most of the cases, the source was operated in the gas discharge mode thus without any chemical selectivity. But for the determination of the  $\alpha$ -branching ratio of the astatine daughters a pure source of the francium mother nuclei was obtained by running the source in the surfaceionization mode, suppressing the ionization of radon and astatine isotopes.

The mass-separated beam was implanted into an aluminized Mylar tape which periodically moved the source from the implantation station to the decay station. The cycle time was adjusted to optimize the counting rate for the nucleus under study. Another experimental setup consisted of a wheel with five  $30 \ \mu g/cm^2$  thick carbon foils. At the decay station, particle detectors on both sides viewed the activity.

Singles  $\gamma$  spectra were taken with two Ge detectors with a resolution of 2.0 keV and an efficiency of 20% at the 1332.5 keV line of <sup>60</sup>Co relative to the efficiency of a 7.6 cm×7.6 cm NaI(Tl) detector. Singles x-ray spectra were accumulated with a LeGe-type (low-energy germanium) detector. This detector has an active surface of 1500 mm<sup>2</sup> and a resolution of 580 eV for the 122 keV line of <sup>57</sup>Co. Intensity calibrated sources were used for energy and intensity calibration of these detectors. A Si(Li) detector (Kevex type, thickness 5mm) was used to detect conversion electrons. For the 624 keV  $e^{-1}$  line of <sup>137</sup>Cs, a resolution of 2.5 keV was obtained. This  $e^{-1}$  detector faced the radioactive source directly at a variable dis-

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tance between 3 and 6 cm. Energy and intensity calibrations were performed using strongly converted transitions with known conversion coefficients. Several  $\alpha$  detectors of the surface barrier type (450 mm<sup>2</sup>, 20 keV resolution on the 5.486 MeV  $\alpha$  line of <sup>241</sup>Am) and of the PIPS type (50–150 mm<sup>2</sup>, 11–17 keV resolution) were used. The efficiency of the  $\alpha$  detectors was calculated from the solid angle. Multiscaled singles spectra were taken with all these detectors in order to obtain half-life information. A restricted set of  $\alpha\gamma t$  and  $\alpha Xt$  triple coincidences was also collected.

#### **III. RESULTS**

## A. The <sup>206</sup>Fr→<sup>202</sup>At→<sup>198</sup>Bi a-decay chain

Ritchie et al. [7] reported the existence of a 0.7 s isomer in <sup>206</sup>Fr decaying with a 6.930 MeV  $\alpha$  line and with a 531 keV IT towards the ground state of <sup>206</sup>Fr. This ground state decays with a half-life of 15.9 s: the  $\alpha$ -decay energy is 6.790 MeV. The  $\alpha$ -decay daughter <sup>202</sup>At is known to decay for 15% by the emission of two  $\alpha$  lines (6.133 and 6.227 MeV) [8]. Only a few  $\gamma$  lines have been observed in the  $\beta^+/EC$  decay of <sup>202</sup>At [9]. As these  $\gamma$ lines have also been observed in more detailed in-beam studies, one can conclude that there is  $\beta^+$ /EC feeding to levels with spin values up to 6 (Ref. [9]). There is a wellknown high spin isomer  $(10^{-})$  in <sup>198</sup>Bi decaying with a 7.7 s E3 IT to a  $(7^+)$  state in <sup>198</sup>Bi [10]. The  $\beta^+$ /EC decay of the  $7^+$  state in <sup>198</sup>Bi has been studied in some detail [10]. In a systematic study to observe low-lying  $0^+$  intruder states in the even-even Pb isotopes [11], we obtained evidence for two  $\beta^+/EC$  decaying states in <sup>198</sup>Bi, with roughly the same half-life. Besides the known  $7^+$  state, a low-spin isomer is also  $\beta^+$ /EC decaying. The production of this isomer can be enhanced via the  $\beta^+$ /EC decay of <sup>198</sup>Po [11].

Figure 1 gives the  $\alpha$  spectrum of continuously implanted <sup>206</sup>Fr, with the ion source in surface-ionization mode. We do confirm the results of Ritchie *et al.* [7]: two  $\alpha$ lines [6.792(5) and 6.930(5) MeV] are attributed to the decay of <sup>206</sup>Fr. The other  $\alpha$  lines are coming from daughters of <sup>206</sup>Fr: the 6.261 MeV line from <sup>206</sup>Rn after  $\beta^+$ /EC decay and the 6.135 and 6.228 MeV lines from  $^{202}$ At after  $\alpha$  decay. The small peak at 5.588 MeV indicates the presence of the granddaughter  $^{202}$ Po, a decay product of  $^{202}$ At and  $^{206}$ Rn. By using the genetic relations and the proper correction procedure [12], it is possible to obtain some  $\alpha$ -branching ratios. We will present the results later on.

Figure 2 gives the sum spectrum at mass 202 of a  $6 \times 90$  s implantation cycle, obtained in the Re(<sup>20</sup>Ne,*xn*) reactions. All  $\alpha$  lines, except the one at 6.277 MeV, are known. The half-life of the 6.277 MeV line is 0.46(5) s, similar to the half-life of a 391.7(2) keV transition present in the  $\gamma$  spectrum at mass 202. This transition is also seen in the electron spectrum and from the electronbinding energy, we could conclude that the 391.7 keV transition occurs in astatine. The conversion coefficient of this isomeric transition has been measured  $[\alpha_K = 8.8(12) \times 10^{-2}$  and  $\alpha_K / \alpha_L = 0.62(1)]$  which fixes the multipolarity to E3. It is now possible to combine the E3 internal transitions in <sup>206</sup>Fr, <sup>202</sup>At, and <sup>198</sup>Bi with four  $\alpha$  lines (6.930 MeV from <sup>206</sup>Fr<sup>m</sup>, 6.792 MeV from <sup>202</sup>At<sup>m</sup>, and 6.135 MeV from <sup>202</sup>At) in a 10<sup>-7+</sup> decay chain (see Fig. 3).

On the basis of half-life measurements, Hornshoj et al. conclude in Ref. [8] that the 6.135 and 6.228 MeV  $\alpha$  lines in <sup>202</sup>At are originating from the same state. However the ratio of the 6.135 to 6.228 MeV  $\alpha$ -line intensity is changing from 2.00(4) to 1.1(1) when going from direct production (see Fig. 1) to production via <sup>206</sup>Fr (see Fig. 2). Schmorak explains this by a  $^{206}$ Rn impurity in the spectra of Hornshoj et al [9]. Careful peak fitting of the spectrum in Fig. 1 rules out this possibility. Furthermore, in a recent search for fine structure in the  $\alpha$  decay of <sup>202</sup>Rn, only the 6.228 MeV line of <sup>202</sup>At was observed as a decay product in the mass-separated and chemically purified source of  $^{202}$ Rn [5]. Clearly a third  $\alpha$ -decaying isomer is present in  ${}^{202}$ At. This isomer has a low spin as it is fed in the  $\beta^+/EC$  decay of the even-even  ${}^{202}$ Rn. It decays to the low-spin isomer in <sup>198</sup>Bi: A similar enhancement of the low-spin state population of <sup>198</sup>Bi as seen in the  $\beta^+$ /EC decay of <sup>198</sup>Po is observed here. The  $4^+ \rightarrow 2^+$  to  $2^+ \rightarrow 0^+$  intensity ratio in <sup>198</sup>Pb is changing from 0.79 (direct production) to 0.50 (via <sup>198</sup>Po) and to

FIG. 1. The  $\alpha$  spectrum of continuously implanted <sup>206</sup>Fr, with the ion source in surface-ionization mode.



FIG. 2. The sum spectrum at mass 202 of a  $6 \times 90$  s implantation cycle.



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FIG. 3. The  ${}^{206}\text{Fr} \rightarrow {}^{202}\text{At} \rightarrow {}^{198}\text{Bi}$  decay chain. Full arrows symbolize  $\alpha$  decay, open arrows  $\beta^+/\text{EC}$  decay, and broken arrows  $\gamma$  decay. The half-lives are given in seconds; the energies in keV. The half-life of the  $10^-$  isomer in  ${}^{206}\text{Fr}$  is from Ref. [7], together with the energy of its IT. The decay data of the  $10^-$  isomer in  ${}^{198}\text{Bi}$  are from Ref. [10].

0.56 (via  $^{202}$ At). As the 6.135 and 6.228 MeV are both seen in the decay of <sup>206</sup>Fr and as there is only one intense  $\alpha$  line in <sup>206</sup>Fr (see Fig. 1), this  $\alpha$  line (6.792 MeV) must be a doublet. The  $\alpha\gamma t$  coincidences on mass 202 show coincidences between a 5.929(10) MeV  $\alpha$  line and a 139(1) and 164(1) keV  $\gamma$  line and between a 6.07(1) MeV  $\alpha$  line and the 164 keV  $\gamma$  line. As the energy of the crossover  $\alpha$  line matches the 6.228 MeV line, we attribute the two  $\alpha$  lines to the decay of the low-spin isomer. Figure 3 summarizes the decay of the three isomers in  $^{206}$ Fr,  $^{202}$ At, and  $^{198}$ Bi. The half-life of the low-spin and high-spin  $\beta^+$ /EC states in <sup>198</sup>Bi has been measured by recording the time behavior of the conversion electrons of respectively the  $0^+$  to  $0^+$  transition and the 7<sup>-</sup> to 5<sup>-</sup>, 5<sup>-</sup> to 4<sup>+</sup>, and 4<sup>+</sup> to 2<sup>+</sup> transitions in <sup>198</sup>Pb. The spin and parities of the different isomers are based on the  $(7^+)$  and  $(2^+, 3^+)$  assignment for spin and parity of the two  $\beta^+$ /EC-decaying states in <sup>198</sup>Bi. This is based on the  $\beta^+$ /EC-decay pattern and on systematics. We used fast  $\alpha$  decay, characterized by small hindrance factors as compared to the even-even neighbors, as a strong rule for a transition without spin and parity changes. The  $\alpha$ -branching ratios and deduced  $\alpha$ -reduced widths will be discussed later.

# B. The <sup>204</sup>Fr-<sup>200</sup>At-<sup>196</sup>Bi decay chain

Hornshoj *et al.* [8] attributed two  $\alpha$  lines to the decay of <sup>204</sup>Fr. The two lines [7.027(5) and 6.967(5) MeV], have the same half-life (2.1 s) and thus, according to Hornshoj, deexcite the same state. Two  $\alpha$ -decaying isomers are known in <sup>200</sup>At [13]. The 43 s isomer decays for 35% by the emission of three  $\alpha$  lines [6.412(2), 6.465(2), and 6.574(5) MeV]. The 4.3 s isomer decays by a 6.536(4) MeV  $\alpha$  line and probably by a  $\simeq$  240 keV E3 internal transition [13]. The decay of <sup>196</sup>Bi has been studied at the LISOL separator [11,14]: a (10<sup>-</sup>) isomeric state decays by  $\alpha$  emission, by  $\beta^+$ /EC, and by a 102 keV E3 IT to a (7<sup>+</sup>) state; the low-spin (3<sup>+</sup>) ground state decays by  $\beta^+$ /EC and by  $\alpha$  emission.

Figure 4 shows the  $\alpha$  spectrum of continuously implanted <sup>204</sup>Fr, mass-separated with the ion source in surface-ionization mode. The known  $\alpha$  lines of <sup>204</sup>Fr are present but the line at 7.027 MeV is a doublet consisting of a  $\simeq 1$  s 7.013 MeV line and a 1.7(3) s 7.031 MeV line. The half-life of the 6.969 MeV line is 2.6(3) s. The  $\alpha\gamma t$ coincidences show a coincidence between a 6.916(8) MeV  $\alpha$  line and a 113(1) keV M1  $\gamma$  transition. The energy sum of these two correspond to the 7.031 MeV  $\alpha$  line. Finally, a  $\alpha$  line at 7.077(8) MeV is visible in the singles  $\alpha$  spectrum of <sup>204</sup>Fr, with the ion source in the Febiad mode. A partial account of our  $\alpha$ -decay study on <sup>200</sup>At has been given in Ref. [11]. The situation is quite similar to the decay of <sup>202</sup>At: again three isomers are present. A 3.5(2) s (10<sup>-</sup>) state decays by  $\alpha$  emission and by a 230.9(2) keV E3 IT to a  $(7^+)$  state. The conversion coefficients of the 230.9 keV line are  $\alpha_K = 0.29(8)$ ,  $\alpha_L = 1.1(2)$ , and  $\alpha_M = 0.27(6)$ . The  $(7^+)$  state at 104 keV above the ground state has a half-life of 47 s and decays by  $\beta^+/EC$ 



FIG. 4. The  $\alpha$  spectrum of continuously implanted <sup>204</sup>Fr, with the ion source in surface-ionization mode.



FIG. 5. The  ${}^{204}\text{Fr} \rightarrow {}^{200}\text{At} \rightarrow {}^{196}\text{Bi}$  decay chain. Full arrows symbolize  $\alpha$  decay, open arrows  $\beta^+$  /EC decay, and broken arrows  $\gamma$  decay. The half-lives are given in seconds; the energies in keV.

and by  $\alpha$  emission. The ground state has a low spin (3<sup>+</sup>) and decays by  $\beta^+$ /EC and  $\alpha$  emission. Figure 5 summarizes the different decay channels of the three isomers in <sup>204</sup>Fr, <sup>200</sup>At, and <sup>196</sup>Bi. All spin and parities are based on the (10<sup>-</sup>) and (3<sup>+</sup>) assignment of the two  $\beta^+$ /ECdecaying states in <sup>196</sup>Bi. This assignment is based on the feeding pattern in the  $\beta^+$ /EC decay and on systematics. Fast  $\alpha$  decay has been taken as an indication for a transition without changes in spin and parity.

## C. The <sup>202</sup>Fr-<sup>198</sup>At-<sup>194</sup>Bi decay chain

Only one  $\alpha$  branch has been attributed to the decay of  $^{202}$ Fr: a 0.34 s  $\alpha$  line of 7.251 MeV [9]. Two  $\alpha$ -decaying states are reported for  $^{198}$ At: a 4.9 s isomer decaying with a 6.748 MeV line and a 1.5 s isomer decaying with a 6.847 MeV line [10]. The  $\beta^+$ /EC and  $\alpha$  decay of  $^{194}$ Bi has been studied at the LISOL separator: a 115(4) s (10<sup>-</sup>) and a 95(3) s 3<sup>+</sup> state are both decaying via  $\beta^+$ /EC and  $\alpha$  emission [11,14]; they are lying closely together but their relative position is not known.

Figure 6 gives the sum of a  $4 \times 2$  s implantation cycle



FIG. 6. The sum  $\alpha$  spectrum of a 4×2 s implantation cycle of mass 202.

of mass 202 with the ion source in the Febiad mode. The one  $\alpha$  line from <sup>202</sup>Fr can be seen; however, the energy determination fixes its energy at 7.237(8) MeV. The  $\alpha$ lines of the two <sup>198</sup>At isomers are also visible. The other lines are mainly from <sup>202</sup>Rn and <sup>202</sup>At. The fact that the two isomers of <sup>198</sup>At are both fed by the same  $\alpha$  line from <sup>202</sup>Fr indicates that this one transition has again a doublet structure and that there are at least two  $\alpha$ -decaying isomers in  $^{202}$ Fr. At mass 198, we do observe the known  $\alpha$ lines of <sup>198</sup>At. In the  $\alpha\gamma t$  coincidences two more  $\alpha$  lines are visible. The 6.360(10) MeV line is coincident with a 181(1) and 218(1) keV  $\gamma$  line; the 6.755 MeV  $\alpha$  line of the 4.2 s isomer of <sup>198</sup>At can be the crossover  $\alpha$  transition by placing the two  $\gamma$  rays in cascade (see Fig. 7). The 6.539(6) MeV  $\alpha$  line has been weakly seen in the  $\alpha\gamma t$ coincidences: Due to low statistics no clear coincidence relation with a  $\gamma$  ray could be obtained but the  $\alpha$  line fits in the decay scheme of the 4.2 s isomer (see Fig. 7). Also one of the daughters of <sup>198</sup>At gave some  $\alpha\gamma t$  coincidences in the mass-198 run: the 5.645 MeV  $\alpha$  line of the 34.6 s 3<sup>+</sup> state of <sup>194</sup>Bi is coincident with a 151 keV transition [14]. The 5.599 MeV  $\alpha$  line of the (10<sup>-</sup>) isomer of <sup>194</sup>Bi is not seen in the  $\alpha Xt$  coincidence data although there is a strong coincidence relation between this line and a 64 keV transition [14]. This makes it possible to relate, on intensity balances, the 5.645 MeV  $\alpha$  line of the 3<sup>+</sup> state in  $^{194}$ Bi to the decay of the 6.755 MeV decay of 4.2 s  $^{198}$ At. The 1.0 s decay of  $^{198}$ At is then feeding the (10<sup>-</sup>) state in



FIG. 7. The <sup>202</sup>Fr $\rightarrow$ <sup>198</sup>At $\rightarrow$ <sup>194</sup>Bi decay chain. Full arrows symbolize  $\alpha$  decay, open arrows  $\beta^+$ /EC decay, and broken arrows  $\gamma$  decay. The half-lives are given in seconds; the energies in keV. The half-lives of the <sup>202</sup>Fr isomers are from Ref. [9].

<sup>194</sup>Bi. No evidence was found for an isomeric transition in <sup>198</sup>At. A summary of the <sup>202</sup>Fr-<sup>198</sup>At-<sup>194</sup>Bi decay chain is given in Fig. 7. All spin and parities are again based on the (10<sup>-</sup>) and 3<sup>+</sup> assignment of the two isomers in <sup>194</sup>Bi. This assignment is based on the feeding pattern in the  $\beta^+$ /EC decay, on systematics, and, for the 3<sup>+</sup> state, on the observation of unhindered  $\alpha$  decay to a 3<sup>+</sup> state in <sup>190</sup>Tl [14]. Fast  $\alpha$  decay has been taken as an indication for a transition without changes in parity and spin.

## **D.** The $\beta^+$ /EC decay of <sup>202</sup>At

Three  $\gamma$  lines have been attributed to the  $\beta^+/\text{EC}$  decay of <sup>202</sup>At. These  $\gamma$  lines have also been seen in in-beam studies and are identified as the three lowest members of the ground-state band [9]. In Table I, we give a complete list of  $\gamma$  lines belonging to the  $\beta^+/\text{EC}$  decay of the two isomers in <sup>202</sup>At. As the difference in half-life of the two isomers (182 and 184 s) is so small, it will not be possible to attribute the  $\gamma$  rays to one of the isomers on the basis of the time behavior. We have not performed a detailed decay study on <sup>202</sup>At, but we can construct a level scheme mainly on the basis of the in-beam work [9,15] and on the basis of the systematics of the neighboring At isotopes. As can be seen in Fig. 8, there is substantial feeding to the known 6<sup>+</sup>, 8<sup>+</sup>, and 9<sup>-</sup> states. On the basis of the systematics of the 5<sup>-</sup>, 7<sup>-</sup>, and 9<sup>-</sup> "neutron" levels in the



FIG. 8. The  $\beta^+$ /EC decay scheme of the 7<sup>+</sup> and 2<sup>+</sup>, 3<sup>+</sup> isomers of <sup>202</sup>At.

TABLE I. List of  $\gamma$  rays attributed to the  $\beta^+$ /EC of <sup>202</sup>At.

Energy (keV)	Intensity (%)	Assignment	
443.4(2)	59(1)	6 <sup>+</sup> -4 <sup>+</sup>	
526.8(2)	3.0(3)	98+	
571.6(2)	88(1)	<b>4</b> <sup>+</sup> - <b>2</b> <sup>+</sup>	
603.3(2)	17.8(3)	(78+)	
617.8(2)	15.4(4)	(54+)	
625.3(3)	$\simeq 1.4$	(76+)	
649.8(2)	5.5(3)		
677.2(2)	100	2+-0+	
755.1(2)	7.3(4)		

even polonium nuclei (see Fig. 10 in Ref. [16]), we propose a candidate for the 5<sup>-</sup> level at 1866.6 keV and for the 7<sup>-</sup> level at 2295.4+ $\Delta$ . These levels have to be confirmed by coincidence measurements. If this construction is correct, the value for  $\Delta$ , the difference between the 8<sup>+</sup> and 6<sup>+</sup> levels, would be 22 keV. Although there is some uncertainty in the placement of some  $\gamma$  rays, one can conclude out of the feeding pattern that the  $\beta^+$ /EC decay is mainly coming from the 7<sup>+</sup> isomer in  $^{202}$ At. At least 73% of the total decay is coming from this 7<sup>+</sup> isomer; this lower limit will further on be used for the determination of the  $\alpha$ -branching ratios of the different isomers in  $^{202}$ At.

## E. The $\beta^+$ /EC decay of <sup>200</sup>At

The  $\beta^+/EC$  decay of <sup>200</sup>At has not been studied yet. However there exists an extensive in-beam study of <sup>200</sup>Po [16]. Table II lists the  $\gamma$  rays attributed to the decay of  $7^+$  and  $3^+$  isomers in <sup>200</sup>At. The difference in half-life (47 and 43 s) is again too small to make any assignment to the different isomers. The multipolarity, based on the conversion coefficients, of some of the strong  $\gamma$  lines has been determined (see Table II). A level scheme has been constructed on the basis of intensity balances, energy sums, the in-beam work [16], and the systematics of the

TABLE II. List of  $\gamma$  rays attributed to the  $\beta^+$ /EC of <sup>200</sup>At.

Energy (keV)	Intensity (%)	Multipolarity	Assignment
264.8(2)	0.6(1)		
323.7(2)	3.2(1)		75-
361.6(2)	0.9(1)		78+
374.0(2)	7.0(1)		76+
409.3(2)	3.3(1)		
484.5(2)	49.8(4)		6 <sup>+</sup> -4 <sup>+</sup>
488.1(2)	1.9(2)		98+
514.8(2)	1.7(1)		
534.5(2)	16.5(2)	E1	54+
565.0(2)	17.0(2)	<i>M</i> 1	$3^+, 4^+-4^+$
611.1(2)	85.0(6)	E2	4+-2+
659.6(3)	0.3(1)		
666.0(3)	100	E2	$2^{+}-0^{+}$
1177.1(5)	1.4(2)		3+,4+-2+

neighboring At isotopes (see Fig. 9). Our level scheme confirms the in-beam work. The ordering of the  $4^+ \rightarrow 2^+$  and  $2^+ \rightarrow 0^+$  transitions in Ref. [16] has in fact been based on private communication of our work. Similar as to the case of <sup>202</sup>At, it is mainly the 7<sup>+</sup> isomer in <sup>200</sup>At which is  $\beta^+$ /EC decaying. This will further on be used for the determination of the  $\alpha$ -branching ratios of the different isomers in <sup>200</sup>At.

#### F. Determination of the branching ratios

Three methods have been used to determine the branching ratios of the different isomers. The first and most reliable one is by comparing the  $\alpha$  decay of the feeding parent nucleus and the daughter nucleus. Massseparated and eventually chemically purified sources are necessary. A proper correction procedure needs to be used [12]. A second method, especially suited for the comparison of  $\alpha$  and IT decay, is to relate the intensity of the  $\alpha$  line with the electron lines of the isomeric transition under study. No half-life nor detector efficiency corrections are necessary as one can measure the  $\alpha$  and electron particles in the same detector. The third method consists of the comparison of the  $\alpha$  decay to the  $\gamma$  decay (after  $\beta^+$ /EC or as IT). Two detectors are used, with the inherent uncertainties of efficiency, counting time, and pile-up corrections. Furthermore, the relation between

the  $\gamma$  and  $\beta^+$ /EC decay has to be known precisely. Table III summarizes the obtained  $\alpha$ -branching ratios and indicates which method has been used. We will not discuss the table in detail. However some values need to be clarified. We have not included the  $\alpha$ -branching ratio of  $^{206}$ Fr<sup>m</sup> from Ref. [7] as it was obtained by comparing the intensities of the 206 Fr<sup>m</sup> and 206 Fr  $\alpha$  groups; this is not a correct procedure as the heavy-ion reaction will not populate only the  $(10^-)$  state but also the  $(7^+)$  and the  $(2^+, 3^+)$  state. The  $\alpha$ -branching ratio of  $^{206}$ Fr is for the  $(7^+)$  and  $(2^+, 3^+)$  isomers together as it is impossible to decompose the doublet. The  $\alpha$ -branching ratio, with the ion source in surface-ionization mode, is obtained by comparing the intensity of the 6.792 MeV line of <sup>206</sup>Fr to the 6.261 MeV line of <sup>206</sup>Rn, the  $\beta^+$ /EC daughter of <sup>206</sup>Fr. The  $\alpha$ -branching ratio of <sup>206</sup>Rn [68(3)%] is taken from Ref. [17]. Our value agrees well with the value from Hornshoj et al. [85(2)%] [8] but differs from the value of Ritchie et al. [93(4)%] [7]. The  $\alpha$ -branching ratio of the  $(2^+, 3^+)$  isomer in <sup>202</sup>At is only a lower limit: This value is obtained by attributing a maximum of 27% of the  $\beta^+/EC$  to the low-spin isomer (see Sec. III D). The lower limits for the  $\alpha$ -branching ratios of the two isomers in <sup>198</sup>At are obtained by comparing the  $\alpha$ -line intensity of the one isomer to the  $^{202}$ Fr  $\alpha$ -line intensity and assuming 100% for the  $\alpha$ -branching ratio of the other isomer and vice versa. The  $\alpha$ -branching ratios of the doubly-odd Bi isotopes have been recently published by our group [14]. These values are used in Table III with one exception:





FIG. 9. The  $\beta^+$ /EC decay scheme of the 7<sup>+</sup> and 3<sup>+</sup> isomers of <sup>200</sup>At.

FIG. 10. The energy difference of the  $vi_{13/2}$  and  $vf_{5/2}$  states in the odd-mass Pb isotopes ( $\bigcirc$ ), compared to the energy of the  $10^- \rightarrow 7^+ E3$  transitions in Fr ( $\triangle$ ), At ( $\times$ ), and Bi ( $\square$ ). The upper part of the figure gives the B(E3) values of the *j*forbidden isomers in Fr ( $\triangle$ ), At ( $\times$ ), and Bi ( $\square$ ).

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The  $\alpha$ -branching ratio of the  $(10^{-})$  isomer in <sup>196</sup>Bi is now decreased from 0.000 38% to 0.0003% as, in Ref. [14], we did not take into account the 21% of the 102 keV isomeric transition.

## **IV. DISCUSSION**

### A. The E3 isomers

Table IV lists the  $10^- \rightarrow 7^+ E3$  transitions known in the odd-odd francium, astatine, and bismuth isotopes, their partial half-life, and their transition strength in Weisskopf units. Figure 10 compares the energy of these E3 transitions to the energy difference between the  $\frac{13}{2}^+(1\nu i_{13/2})$  and  $\frac{5}{2}^-(2\nu f_{5/2})$  neutron states in the odd lead isotopes. There is a remarkable similarity in the energy behavior as a function of the neutron number. Furthermore, as can be seen in Table IV, the E3 transitions are highly retarded. They are  $10^5$  times slower than other E3 transitions in the neighborhood [18]. Hagemann et al. [19] and Gippner et al. [20] interpreted these slow E3 transitions are  $[\pi(h_{9/2})_{9/2}^n \otimes v(i_{13/2})]_{10}^-$  and  $[\pi(h_{9/2})_{9/2}^n \otimes v(f_{5/2})]_{7^+}$ . The similarity in energy behavior of the  $10^- \rightarrow 7^+$  transition in the bismuth isotopes, relative to the  $\frac{13}{2}^+ \rightarrow \frac{5}{2}^-$  transition in the lead isotones, indicates that the odd proton in bismuth acts as a spectator and that the coupling between the odd proton and the odd neutron stays the same although the number of neutrons is changing from 114 to 125. Adding two or four

TABLE III. A list of  $\alpha$ -decay energies, half-lives,  $\alpha$ -branching ratios, and  $\alpha$ -reduced widths of the nuclei under study. The fine structure in the  $\alpha$  decay of <sup>194</sup>Bi is not listed (see Ref. [14]). Values from other work are indicated by their reference number. The methods used are (1) comparing the  $\alpha$  activity of the mother and the daughter, (2) comparing in one detector the  $\alpha$  to electron activity, and (3) comparing in two detectors the  $\alpha$  to  $\gamma$  activity.

<b>-</b> .		<i>T</i> <sub>1/2</sub>	$E_{\alpha}$	a <sub>B</sub>	$\delta^2$	Method
Isotope	<u> </u>	(s)	(MeV)	(%)	(keV)	
<sup>206</sup> Fr	(10 <sup>-</sup> )	0.7(1) <sup>a</sup>	6.930(5)	≤ 100	≤290	
	$(7^+)$ $(2^+, 3^+)$	15.9(3) <sup>a</sup>	6.792(5)	84(2)	31(1)	1
<sup>204</sup> Fr	(10 <sup>-</sup> )	$\simeq 1s$	7.013(5)	≤ 100	≤93	
	(7+)	2.6(3)	6.969(5)	≤100	≤ 58	
			7.077(8)	$\leq 0.7(2)$	≤0.2	
	(3+)	1.7(3)	7.031(5)	≤ 100	≤ 55	
			6.916(8)	$\leq$ 0.6(2)	0.8	
<sup>202</sup> Fr	(10 <sup>-</sup> ) (3 <sup>+</sup> )	0.34(4) <sup>b</sup>	7.237(8)	≤ 100	≤ 53	
<sup>202</sup> At	(10 <sup>-</sup> )	0.46(5)	6.277(5)	$9.6(1.1) \times 10^{-2}$	21(3)	2.3
	( <b>7</b> <sup>+</sup> )	182(2)	6.135(2) <sup>b</sup>	8.7(1.5)	20(3)	1.2.3
	$(2^+, 3^+)$	184(1)	6.228(2) <sup>b</sup>	$13 \rightarrow 100$	12→89	1.3
			6.070(10)	0.03→0.2	0.1→0.9	-,-
			5.929(10)	$5 \times 10^{-3} \rightarrow 4 \times 10^{-2}$	0.1→0.8	
<sup>200</sup> At	(10 <sup>-</sup> )	3.5(2)	6.538(3)	10.5(3)	28(2)	1,2,3
	(7+)	47(1)	6.411(2)	43(7)	28(5)	1,3
			6.575(3)	0.36(6)	0.05(1)	
			6.306(5)	0.073(15)	0.13(3)	
	(3+)	43(1)	6.464(2)	57(6)	24(3)	1,3
<sup>198</sup> At	(10 <sup>-</sup> )	1.0(2)	6.856(4)	67→100	$37 \rightarrow 73$	1
	(3 <sup>+</sup> )	4.2(3)	6.755(4)	$80 \rightarrow 100$	$26 \rightarrow 37$	1
			6.539(10)	?	?	-
			6.360(10)	$0.28 \rightarrow 0.35$	3.5→4.4	
<sup>198</sup> Bi	$(10^{-})$	7.7°				
	$(7^{+})$	693(18)				
	(2+,3+)	618(20)				
<sup>196</sup> Bi	(10 <sup>-</sup> )	240(3)	5.112(5)	0.0003(1)	8(3)	3
	(7+)	0.6(5)				
	(3+)	308(12)	5.153(5)	0.00115(34)	15(4)	3
<sup>194</sup> Bi	(10 <sup>-</sup> )	115(4)	5.599(5)	0.20(7)	33(12)	3
	3+	95(3)	5.645(5)	0.46(25)	54(29)	3

\*Reference [7].

<sup>b</sup>Reference [9].

<sup>c</sup>Reference [10].

TABLE IV. A list of the  $10^- \rightarrow 7^+ E3$  isomers in the doubly odd Fr, At, and Bi isotopes. The partial half-life is calculated using the formula  $T_{1/2}^{\text{partial}} = T_{1/2}(1 + \alpha_T)/B$ , with  $\alpha_T$  the total conversion coefficient and B the branching ratio. The experimental B(E3) values [in Weisskopf units (W.u.)] are calculated using the formula  $B(E3) = 0.02044 [T_{1/2}^{\text{partial}}(s)E_{\gamma}^{T}(\text{MeV})A^2]^{-1}$ , with  $E_{\gamma}$  the transition energy and A the mass number. This table is based on Ref. [17] and on our work.

	$E_{\gamma}$ (MeV)	T <sub>1/2</sub> (s)	$T_{1/2}^{\text{partial}}$ (s)	<i>B</i> ( <i>E</i> 3) (W.u.)
<sup>206</sup> Fr	0.531	0.7	0.78	$(5.2 \times 10^{-5})$
<sup>204</sup> At	0.587	0.108	0.116	$1.8 \times 10^{-3}$
$^{202}$ At	0.392	0.46	0.59	$6.0 \times 10^{-4}$
<sup>200</sup> At	0.231	3.5	13.9	$1.0 \times 10^{-3}$
<sup>208</sup> Bi	0.921	$2.53 \times 10^{-3}$	$2.53 \times 10^{-3}$	$3.3 \times 10^{-4}$
<sup>206</sup> Bi	0.904	$8.9 \times 10^{-4}$	$7.42 \times 10^{-3}$	$1.3 \times 10^{-4}$
<sup>204</sup> Bi	0.752	$1.3 \times 10^{-2}$	$1.3 \times 10^{-2}$	$2.8 \times 10^{-4}$
<sup>202</sup> Bi	0.598			
<sup>200</sup> Bi	0.428	0.4	0.47	$4.1 \ 10^{-4}$
<sup>198</sup> Bi	0.248	7.7	19.9	$4.5 \ 10^{-4}$
<sup>196</sup> Bi	0.102	240	$\simeq 1.8 \ 10^5$	$\simeq 2.5 \ 10^{-5}$

protons, for astatine and francium, respectively, is not changing this picture. The B(E3) values also do not change significantly by varying the neutron or proton numbers: they are highly retarded due to the *j* forbiddeness ( $\Delta i = 4 > L = 3$ ). There is no evidence for an admixture of fast single-particle transitions of the type  $\pi i_{13/2} \rightarrow \pi f_{7/2}$  or  $\nu j_{15/2} \rightarrow \nu g_{9/2}$  or for collective octupole admixtures [3,18]. It is not clear if the same structure is still present in <sup>204</sup>Fr. As can be seen in Fig. 5 and Fig. 10, the energy difference between the  $(10^{-})$  and  $(7^{+})$  isomers in <sup>204</sup>Fr is 275 keV. This value deviates strongly from the energy systematics of Fig. 10. Further experiments, such as a search for the connecting E3 transition of 275 keV, are necessary to clarify this situation. The B(E3) values for <sup>206</sup>Fr and <sup>196</sup>Bi seem to be extremely low. In <sup>206</sup>Fr the situation is still a bit confusing as the multipolarity and the branching ratio of the transition is not yet measured. In <sup>196</sup>Bi, the B(E3) value depends strongly on the energy, conversion coefficient, and branching ratio of the 102 keV transition, all quantities only roughly known [11,14].

### B. The $\alpha$ -reduced widths

The last column of Table III gives the  $\alpha$ -reduced widths of the main  $\alpha$  transitions in the different nuclei. The  $\alpha$ -reduced widths are calculated using the method of Rasmussen [21] assuming L=0 transitions. For most of the Fr isotopes, the  $\alpha$ -branching ratio is not known and an upper limit of 100% is used. The obtained  $\alpha$ -reduced widths are lying between 10 and 100 keV, a value which is also found in the neighboring even-even isotopes [5,22]. This is a strong fingerprint for unhindered  $\alpha$  decay, thus connecting states with identical spin and parity. A  $\Delta L = 1$  transition within a similar proton-neutron multiplet is immediately hindered by at least a factor of 40 [14].

### V. CONCLUSION

We have presented evidence for the existence of different isomers in the doubly odd, neutron-deficient francium, astatine, and bismuth isotopes. These isomeric states are formed by the coupling of a  $h_{9/2}$  proton with a  $i_{13/2}$  neutron leading to a 10<sup>-</sup> state and the coupling of a  $h_{9/2}$  proton with a  $f_{5/2}$  neutron leading to a 7<sup>+</sup> and a 2<sup>+</sup> or  $3^+$  state. A striking feature is the similarity of the decay characteristics of the different isomers in one nucleus: there are a number of doublets exhibiting a similar halflife. A general conclusion is that the shell-model character of the isomeric states is rather pure; the influence of collective quadrupole and octupole degrees of freedom is negligible. This can be seen in the systematics of the energy difference between the  $\left[\pi(h_{9/2})_{9/2}^n \otimes \nu(i_{13/2})\right]_{10^-}$  and  $[\pi(h_{9/2})_{9/2}^n \otimes \nu(f_{5/2})]_{7^+}$  states in function of the proton and neutron number and also in the B(E3) values of the connecting E3 transition. The E3 transition is highly retarded due to the *j* forbiddeness. Finally all the isomers in the Fr-At-Bi decay chain decay partially by fast  $\alpha$  decay: this is a strong fingerprint for  $\Delta L = 0$  decay between identical members in the involved proton-neutron multiplet.

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