# Fine structure in the $\alpha$ -decay of radium isotopes with mass numbers 209–212

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**Abstract.** Radium isotopes with mass numbers from 209 to 212 have been produced by heavy-ion fusion reactions  $^{204}$ Pb( $^{12}$ C, xn) $^{216-x}$ Ra. Radioactive decay properties were investigated by means of  $\alpha$ - and  $\alpha$ - $\gamma$ -spectroscopy after in-flight separation of the evaporation residues from the projectile beam by the velocity filter SHIP and implantation into a 16-strip position-sensitive Si detector. For the even-even nuclei  $^{210,212}$ Ra we identified  $\alpha$  transitions into the first-excited 2<sup>+</sup>-state of the daughter nuclei  $^{206,208}$ Rn. Weak  $\alpha$  transitions into excited levels of the odd-A daughter nuclei  $^{205,207}$ Rn were also observed.

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#### 1 Introduction

Detailed spectroscopic data are rather scarce for isotopes below the neutron shell at N = 126 of elements above francium. Although most of these isotopes were firstly synthesized and identified more than twenty years ago [1], only few properties such as the energies of the strongest  $\alpha$  transitions and the half-lives are known in most cases. More detailed investigations were performed only for  $^{213}$ Ra [2] and <sup>212</sup>Ra [3]. In our recent study of  $\alpha$ - and  $\gamma$ -decay properties of neutron-deficient isotopes of elements between radon and uranium, new data were collected [4]. The identification of a weak  $\alpha$ -decay branch of <sup>214</sup>Ra into the firstexcited 2<sup>+</sup>-state of <sup>210</sup>Rn lead us also to search for  $\alpha$  transitions from the lighter even-even isotopes  $^{210,212}\mathrm{Ra}$  into the first-excited  $2^{+}$ -states in  $^{206,208}$ Rn and to determine their intensities. The energies of these levels are known from the literature [1]. Moreover, it seemed worthwhile to investigate extensively the  $\alpha$ -decay of the neighbouring odd-mass nuclei <sup>209,211</sup>Ra. Alpha transitions into lowlying levels in the daughter nuclei  $^{205,207}\mathrm{Rn}$  that decay to the ground state by emission of  $\gamma$ -rays or conversion electrons deliver valuable information on the nuclear structure of these nuclei at low excitation energies. So far, only little has been reported about excited levels in <sup>205,207</sup>Rn. Difficult in the investigation of weak  $\alpha$  transitions is that the  $\alpha$ -lines are often covered by more intense lines of lighter isotopes. It has been shown, however, that the measurement of  $\alpha$ - $\gamma$  coincidences after in-flight separation of the

evaporation residues from the primary beam is a powerful tool to identify weak transitions even in the presence of a "background" from decays of other isotopes of up to  $10^4$  times higher intensities.

For the production of these isotopes we chose the reaction  $^{204}$ Pb $(^{12}$ C,  $xn)^{216-x}$ Ra (x = 4-7). According to calculations using the fusion-evaporation code HIVAP [5], sufficiently high cross-sections of (1-100) mb were expected.

#### 2 Experiment

The experiments were performed at GSI, Darmstadt, using a <sup>12</sup>C beam delivered by the high charge state injector with ECR-ion source of the UNILAC accelerator. Beam intensities and energies were  $(0.6-1.2) \times 10^{12}$  ions/s ( $\approx (100-200)$  pnA) and (78–110) MeV, respectively. The targets of <sup>204</sup>Pb (isotopic enrichment 99.73%) with thicknesses of  $\approx 450 \ \mu g/cm^2$  were evaporated on carbon layers of 40  $\ \mu g/cm^2$  (upstream) and covered with 5  $\ \mu g/cm^2$  carbon (downstream). They were mounted on a wheel which rotated synchronously to the beam macro structure [6] (5.4 ms wide pulses at 50 Hz repetition frequency).

The evaporation residues recoiling from the targets with energies of  $\approx 5$  MeV were separated from the primary beam by the velocity filter SHIP [7]. In the focal plane of SHIP they were implanted into a position-sensitive 16strip PIPS detector with an active area of  $(80 \times 35)$  mm<sup>2</sup> ("stop detector") [8]. The kinetic energies of the residues were measured as well as subsequent  $\alpha$ -decays. Since our

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**Fig. 1.** Alpha spectra from the decay of isotopes produced in the reaction  ${}^{12}\text{C} + {}^{204}\text{Pb}$  at bombarding energies of  $E_{\text{lab}} = 78 \text{ MeV}$  (a) and  $E_{\text{lab}} = 103 \text{ MeV}$  (b).

**Table 1.** Energies and intensities of transitions used for  $\alpha$ - $\gamma$  efficiency estimation. Data for <sup>211</sup>Po, <sup>213</sup>Ra were taken from [1], <sup>214</sup>Ra from [4], and <sup>216</sup>Ac from this experiment.

Isotope	$E_{\alpha}/\mathrm{keV}$	$i_{lpha}$	$E_{\gamma}/\mathrm{keV}$	$\epsilon_{\alpha-\gamma}$
<sup>211</sup> Po	$\begin{array}{c} 6568 \\ 6891 \end{array}$	$\begin{array}{c} 0.00544 \\ 0.00557 \end{array}$	$897.8 \\ 569.7$	$\begin{array}{c} 0.023\\ 0.038\end{array}$
$^{214}$ Ra	6505	0.002	641.9	0.036
$^{213}$ Ra	6624	0.49	110.1	0.06
<sup>216</sup> Ac	8188 8271		937.9, 777.5 853.5, 770.9, 694.5	$\begin{array}{c} 0.025\\ 0.034\end{array}$

data acquisition rate was limited to  $\approx 700$  events per second due to the dead time of the system, the beam current was reduced to (100–200) pnA, depending on the production cross-sections. The data rate was composed of one third of the signals being due to implantation of evaporation residues, one third to the  $\alpha$ -decay of these residues, and one third to the  $\alpha$ - or electron-capture decays of daughter products. The background of projectiles or scattered target nuclei was negligible. Operated at a temperature of 258 K, the energy resolution for individual strips was (18–20) keV (FWHM). Summing all strips in the off-line data analysis we obtained typical "stop detector" resolutions of  $\Delta E = (20{-}24)$  keV. Typical  $\alpha$  spectra are shown in fig. 1.

Alpha calibration was performed using the literature values of known isotopes which were also produced in the reactions and implanted into the detector (see figs. 1a and b). Although part of the recoil energy transferred by the  $\alpha$ -particle to the residual nucleus contributes to the pulse height, this method is more accurate than using  $\alpha$ -particles from external sources which lose energy in the dead layer of the detector. Using external sources for calibration one obtains for implanted nuclei  $\alpha$  energies which are typically (40–70) keV too high.

Since the evaporation residues were implanted close to the detector surface, the  $\alpha$ -lines had long tails towards lower energies (figs. 1a, b). As a consequence, weak  $\alpha$ -lines in this energy range were difficult to detect.

Coincidences between  $\alpha$ -particles and  $\gamma$ -rays were measured with a high-purity Ge detector mounted directly behind the "stop-detector". Due to the high single  $\gamma$ -rate of several thousand events per second, only  $\gamma$ -events in coincidence with a signal in the "stop detector" were recorded. The  $\gamma$  detector was calibrated with standard energies from <sup>152</sup>Eu and <sup>133</sup>Ba sources.

An estimate of the  $\alpha$ - $\gamma$  coincidence efficiency was performed using known coincident  $\alpha$  and  $\gamma$  transitions of isotopes produced in this experiment (<sup>204</sup>Pb target) and in additional irradiations of <sup>209</sup>Bi or <sup>208</sup>Pb targets. The obtained results are given in table 1. However, the precision of this procedure was limited due to the following reasons: a) The  $\alpha$  transitions in <sup>211</sup>Po and <sup>214</sup>Ra coincident with  $\gamma$ -rays (see table 1) are very weak and the  $\alpha$ -lines were covered by much stronger lines of other isotopes. Therefore, their  $\alpha\text{-decay}$  rates were calculated from the rates of the main lines using literature values for the relative intensities. For <sup>214</sup>Ra the intensity of the 6505 keV line is only known roughly [4], while for <sup>211</sup>Po only a small number of  $\alpha$ - $\gamma$  coincidences ( $\approx 50$ ) were observed for each transition. b) For <sup>216</sup>Ac the  $\alpha$ -lines are coincident with several  $\gamma$ -lines (see table 1), so only an average value could be used. c) For <sup>213</sup>Ra the intensities had to be corrected for internal conversion. We used the values given in ref. [9] for E2 transitions. Taking into account the uncertainties discussed, we concluded the  $\alpha$ - $\gamma$  coincidence efficiencies are reliable only within a factor of two.

The accuracy of the relative intensities of the  $\alpha$  transitions into excited levels is limited due to the following reasons: a) The absolute efficiency for  $\alpha$ - $\gamma$  coincidences was only roughly known (see discussion before). b) Due to almost identical ground-state  $\alpha$ -decay energies of <sup>212</sup>Ra and <sup>211</sup>Ra, as well as of <sup>210</sup>Ra and <sup>209</sup>Ra, the ground-state transition rate for the individual isotopes is not well established. To correct for this, we used the cross-section ratios obtained from the HIVAP calculations. c) For the low-energy transitions internal conversion has to be considered. In the case of *L*-conversion, energy summing of conversion electrons and  $\alpha$ -particles leads to a sum energy equal to the ground-state transition energy within the detector resolution and thus seems to increase the intensity of the ground-state transition [10]. Since K/L ratios



Isotope	$\sigma_{ m max,exp}/ m mb \ (E_{ m lab}/ m MeV)$	$\sigma_{ m max,HIVAP}/ m mb$ $(E_{ m lab}/ m MeV)$	$E_{\alpha}/\mathrm{keV}$	$Q_{lpha}/{ m keV}$	$i_{lpha}$	$E_{\gamma}/\mathrm{keV}$	Lite $E_{\alpha}/\mathrm{keV}$	erature $T_{1/2}/s$
$^{212}$ Ra	(25) $(79)$	100 (75)		$\begin{array}{c} 7031\pm5\\ 6390\pm5\end{array}$	$0.999 \\ 5 \times 10^{-4}$	$635.1\pm0.2$	$6899.2 \pm 1$	$7  13 \pm 2 \ [1]$
<sup>211</sup> Ra	13 (83)	28 (86)	$\begin{array}{c} 6907\pm 5\\ 6788\pm 5\\ (6647\pm 5)\\ 6627\pm 5\\ 6320\pm 10\\ 6315\pm 10\\ 6255\pm 5\end{array}$	$\begin{array}{c} 7040 \pm 5 \\ 6919 \pm 5 \end{array}$ $\begin{array}{c} 6755 \pm 5 \\ 6442 \pm 5 \\ 6437 \pm 5 \\ 6376 \pm 5 \end{array}$	$\begin{array}{c} 0.99\\ 1\times 10^{-2}\\ 8\times 10^{-4}\\ 7\times 10^{-5}\\ 4\times 10^{-4}\\ 6\times 10^{-4}\end{array}$	$\begin{array}{c} 120.0\pm0.1\\ 162.9\pm0.1\\ 283.0\pm0.1\\ 596.1\pm0.3\\ 601.6\pm0.3\\ 665.0\pm0.1 \end{array}$	$6910 \pm 5$	$13 \pm 2$ [1]
$^{210}$ Ra	3.6 (98)	5.8 (98)	$\begin{array}{c} 7003\pm10\\ 6447\pm5 \end{array}$	$7139 \pm 10 \\ 6572 \pm 5$	$0.999 \\ 3 \times 10^{-4}$	$574.9\pm0.1$	$7019\pm5$	$3.7 \pm 0.2$ [1]
<sup>209</sup> Ra	1 (108)	0.9 (112)	$   \begin{array}{r}     7003 \pm 10 \\     6625 \pm 5 \\     6376 \pm 10   \end{array} $	$7140 \pm 10$ $6754 \pm 5$ $6500 \pm 5$	$5 \times 10^{-3}$ $2 \times 10^{-3}$	$387.0 \pm 0.5$ $633.7 \pm 1.1$	$7008 \pm 5$	$4.6 \pm 0.2$ [1]

**Table 2.** Compilation of measured decay data. The uncertainty of the relative intensities of  $\alpha$  transitions into excited states amounts to a factor of three.  $Q_{\alpha} = (1 + m_{\alpha}/m_d) \times E_{\alpha}$ .

depend sensitively on the transition energies and on the multipolarity of the  $\gamma$ -radiation and the latter one can only be estimated, corrections are very uncertain. Therefore, we estimate the intensities of  $\alpha$  transitions to excited daughter levels to be reliable only within a factor of three.

The measured energies and the relative intensities of the  $\alpha$  transitions of the investigated radium isotopes are listed in table 2. For comparison of transition energies in the cases of  $\alpha$ - $\gamma$  coincidence measurements we prefer to use the  $Q_{\alpha}$ -value.

## 3 Results and discussion

#### 3.1 Cross-sections

Production cross-sections for the isotopes of interest in principle could be obtained from the observed  $\alpha$  rates. In practice, however, the accuracy was limited by two reasons. Firstly, the isotopes  $^{211,212}$ Ra and  $^{209,210}$ Ra have similar  $\alpha$ -decay energies for the most intense lines (see table 2). So, from the count rates only the sum values are obtained. Therefore, the cross-sections for the individual isotopes have been determined by correcting the sum value by the ratio of cross-sections obtained from HIVAP calculations for the individual isotopes. Secondly, the angular distribution of the low energetic evaporation residues at the entrance of SHIP is expected to be extremely broad as a result of a large widening due to neutron emission and scattering in the target and converter foil. According to model calculations [11], SHIP transmission values of  $\approx 0.013$  are expected, which, however, may be reliable only within a factor of three. Our experimental maximum values obtained on this basis are listed in table 2, where also the results of the HIVAP calculations are shown. The bombarding energies are given in brackets. Although the

agreement is quite good, it might be accidential. In the investigation of  $^{212}$ Ra our lowest-energy point was about 3 MeV above the value for the maximum cross-section according to HIVAP calculations. The energy was set by using a 5.1 mg/cm<sup>2</sup> nickel foil in front of the target to degrade the accelerator energy of 85 MeV. The foil, however, turned out to be damaged after the end of the irradiation. Therefore, the value for  $^{212}$ Ra must be regarded as somewhat obscure and is given in brackets.

## 3.2 Isotope <sup>212</sup>Ra

Decay data of <sup>212</sup>Ra were first published by Valli *et al.* [12] who used the reactions <sup>197</sup>Au(<sup>19</sup>F, 4n)<sup>212</sup>Ra and <sup>206</sup>Pb(<sup>12</sup>C, 6n)<sup>212</sup>Ra for synthesis. They reported an  $\alpha$ -decay energy and a half-life of (6869 ± 5) keV and (13 ± 2) s. These values are somewhat lower than the unpublished values of Griffioen and Macfarlane, who obtained 6.90 MeV and 18 s. An energy close to the value of Griffioen and Mcfarlane was reported by Hornshøj *et al.* [13], who produced <sup>212</sup>Ra by spallation of thorium and obtained (6902 ± 5) keV and (13.0 ± 0.2) s. In data tables [1] presently values of (6899.2 ± 1.7) keV and (13.0 ± 0.2) s are adopted. Nuclear structure of the  $\alpha$ -decay daughter <sup>208</sup>Rn has been investigated by several groups either using in-beam  $\gamma$ -spectroscopy [14–16] or electron capture and positron decay of <sup>208</sup>Fr [17]. For the first 2<sup>+</sup>-level an energy of 635.8 keV was obtained.

In our experiment we identified an  $\alpha$ -decay branch of <sup>212</sup>Ra into the first 2<sup>+</sup>-level of <sup>208</sup>Rn by coincidences between  $\gamma$  transitions of (635.1±0.2) keV and  $\alpha$ -particles of (6269±5) keV. The relative intensity of this transition is  $\approx 0.0005$ , which is roughly a factor of three lower than the value 0.0016 expected from theoretical  $\alpha$  half-lives including spin hindrance [18,19]. The result of our



Fig. 2. Plots of  $\alpha$ - $\gamma$  coincidences observed for <sup>12</sup>C + <sup>204</sup>Pb at bombarding energies of  $E_{lab} = 78$  MeV (a) and  $E_{lab} = 103$  MeV (b). The accumulation of points in a) is assigned to coincidences of the 6269 keV  $\alpha$  transition with a  $\gamma$  transition of 635.1 keV and in b) to coincidences of the 6447 keV  $\alpha$  transition with a  $\gamma$  transition of 574.9 keV. The decay schemes for <sup>212</sup>Ra and <sup>210</sup>Ra are inserted at the right-hand side. The energies are given in keV and refer to the  $Q_{\alpha}$ -values.

measurement is shown in fig. 2a. It should be stressed out that at the energy of the  $\alpha$  transition strong lines of  $^{212,206,205}$ Rn and  $^{212}$ Fr were present in the single spectra, which made a direct observation of this weak  $\alpha$  fine-structure line impossible.

#### 3.3 Isotope <sup>211</sup>Ra

The isotope <sup>211</sup>Ra has been first synthesized and identified by Valli *et al.* [12] using the reactions <sup>197</sup>Au(<sup>19</sup>F, 5n)<sup>211</sup>Ra and <sup>206</sup>Pb(<sup>12</sup>C, 7n)<sup>211</sup>Ra. They reported an  $\alpha$  energy and a half-life of (6910 ± 5) keV and (15 ± 2) s. There is only scarce information on the nuclear structure of the  $\alpha$ -decay daughter <sup>207</sup>Rn. Rezanka *et al.* [20] reported a 13/2<sup>+</sup> isomeric state at  $E^* = 899$  keV with a half-life of 181  $\mu$ s, decaying to the ground state via a level at 665.1 keV with assumed spin and parity of 9/2<sup>-</sup>.

In our experiment we observed five groups of  $\alpha$ - $\gamma$  coincidences which were attributed to the decay of <sup>211</sup>Ra due to the agreement of  $Q_{\alpha} + E_{\gamma}$  with the value  $Q_{\alpha}(\text{gs})$ = 7040 keV of the line attributed to the ground-state–to– ground-state transition (table 2, figs. 3 and 4). It should be stressed out that the 665.0 keV line perfectly agrees with the energy of the line observed by Rezanka *et al.* from the decay of the 13/2<sup>+</sup> isomeric state in <sup>207</sup>Rn. A sixth group ends up with  $Q_{\alpha} + E_{\gamma} = (6938 \pm 5) \text{ keV}$ , *i.e.* in an about 100 keV lower value. It is also attributed to the decay of <sup>211</sup>Ra, since the coincident  $\gamma$  energy of 162.9 keV is exactly the difference between the 283.0 keV and 120.0 keV lines. Therefore, this transition is assigned to the first step of a two-step decay of the 283.0 keV level.

Information on the multipolarity of the 120.0 keV transition is obtained from the intensity ratio of  $\gamma$ - and X-ray events. Using a fluorescence yield of  $\omega = 0.967$  [1] for radon and assuming equal detection efficiencies for  $\gamma$ - and



Fig. 3. Spectrum of  $\gamma$  events coincident to  $\alpha$ -particles fulfilling the condition 6850 keV  $\langle E_{\alpha} + E_{\gamma} \rangle \langle 6950$  keV. For  $\gamma$  events in the range (160–165) keV we require 6750 keV  $\langle E_{\alpha} + E_{\gamma} \rangle \langle 6950$  keV. The accumulations of events at (200–280) keV and (310–400) keV are due to random coincidences with  $\alpha$  decays of <sup>210,211</sup>Fr (6543 keV, 6535 keV) and <sup>209</sup>Fr (6648 keV). The peak indicated at about 115 keV has a width of 5.1 keV (FWHM) which is considerably broader than that of the 120 keV  $\gamma$ -line (1.4 keV (FWHM)). It is not regarded as due to a  $\gamma$  transition, but as an accumulation of chance coincidences.

X-rays, we obtain a conversion coefficient of  $\alpha = 4.5 \pm 0.5$ which is comparable with  $\alpha = 6.7$  expected for an M1transition according to [9]. The reliability of this method has been tested by application to  $\alpha$ - $\gamma$  coincidences of <sup>213</sup>Ra which was produced in the same reaction. The multipolarity of the 110 keV  $\gamma$ -line emitted in coincidence with  $\alpha$ -particles of 6624 keV has been determined as E2 [2]. From the rates of X-rays and the 110 keV line we obtain  $\alpha = 0.41 \pm 0.02$  which is close to the theoretical value of  $\alpha = 0.38$  [9]. Therefore we conclude that in the case of <sup>211</sup>Ra the 120 keV  $\gamma$  transition is not pure



Fig. 4. Suggested decay scheme for <sup>211</sup>Ra. Energies refer to the  $Q_{\alpha}$ -values and are given in keV. The intensities are given in percent.

*M*1, but has an admixture of *E*2. Using the relation  $\alpha_{\exp} = \delta \times \alpha(E2) + (1 - \delta) \times \alpha(M1)$ , we obtain an admixture coefficient  $\delta \approx 0.4$ . The suggested decay scheme is shown in fig. 4.

In the neighbouring N = 121 isotones <sup>205</sup>Po and <sup>203</sup>Pb, two low-lying levels with energies and spins of 143.166 keV,  $1/2^{-}$  and 154.195 keV,  $3/2^{-}$  (<sup>205</sup>Po) and 126.51 keV,  $1/2^{-}$  and 186.50 keV,  $3/2^{-}$  (<sup>203</sup>Pb) are known from the literature [1]. Due to the multipolarity of the  $\gamma$ radiation we assign the 120.0 keV level in  $^{207}$ Rn to the corresponding  $3/2^{-}$ -level. However, it is remarkable that we do not observe  $\alpha$ -decay into a  $1/2^{-}$ -level located close to the  $3/2^{-}$ -level, while  $\alpha$ -decay of the isotonic nucleus <sup>209</sup>Rn feeds both levels in <sup>205</sup>Po with an intensity ratio of  $i(1/2^{-})/i(3/2^{-}) \approx 0.65$  [1]. The decay  $1/2^{-} \rightarrow 5/2^{-}$ (ground state) is an E2 transition, while the decay  $3/2^{-1}$  $\rightarrow 5/2^-$  (ground state) is an M1 transition. According to [9], we expect a ratio of the total conversion coefficients  $\alpha_{\rm tot}(M1)/\alpha_{\rm tot}(E2) \approx 2.1 \ (\alpha_{\rm tot} = \alpha_K + \alpha_L + \alpha_M).$  Therefore, the  $\alpha$ - $\gamma$  coincidence rate should be about 50% higher for the E2 transition. This estimate is in-line with our result  $i[\gamma(1/2^- \rightarrow 5/2^-)]/i[\gamma(3/2^- \rightarrow 5/2^-)] = 2.5^{+2.5}_{-1.3}$ from the decay of  $^{209}$ Rn, which was produced by  $\alpha$ -decay of <sup>213</sup>Ra in this experiment.

Provided that  $\alpha$ -decay rates into both daughter levels and their excitation energies are similar for the decay <sup>211</sup>Ra $_{\rightarrow}^{\alpha}$  <sup>207</sup>Rn, we should have observed clearly  $\gamma$ -rays from the transition  $1/2^- \rightarrow 5/2^-$  in coincidence with  $\alpha$ -particles. We registered about 600  $\alpha$ - $\gamma$  coincidence events from the decay of the  $3/2^-$ -level and even if we respect an E2 admixture (see above) that lowers the conversion rate, several hundred  $\alpha$ - $\gamma$  coincidence events from the decay of the  $1/2^-$ -level could be expected. Thus, we conclude that a  $1/2^-$ -level in <sup>207</sup>Rn close to the  $3/2^-$ -level (if it exists at all) is populated with an intensity more than an order of magnitude lower than that for the  $3/2^-$ -level by the  $\alpha$ -decay of <sup>211</sup>Ra. A comparison of the level schemes for the N = 121 isotones <sup>203</sup>Pb, <sup>205</sup>Po, <sup>207</sup>Rn (see fig. 5) shows that the energy of the  $1/2^-$ -level increases by 16.6 keV from <sup>203</sup>Pb to <sup>205</sup>Po, while the energy of the  $3/2^-$ -level decreases by 32.3 keV. A decrease by 34.2 keV from <sup>205</sup>Po



**Fig. 5.** Comparison of low-lying levels of the N = 121 isotones <sup>203</sup>Pb, <sup>205</sup>Po, <sup>207</sup>Rn. Energies are given in keV.

to <sup>207</sup>Rn seems reasonable, thus supporting our spin and parity assignment of  $3/2^-$  to the 120.0 keV level in  $^{207}$ Rn. According to the energy difference of the  $1/2^{-}$ -levels in  $^{203}$ Pb and  $^{205}$ Po, we could expect this level in  $^{207}$ Rn at  $E \approx 160 \text{ keV}$ , which is indeed close to 162.9 keV measured for the coincident  $\gamma$  transition. However, it cannot be attributed to the  $1/2^- \rightarrow 5/2^-$  transition, since, as pointed out above, the sum  $Q_{\alpha} + E_{\gamma}$  is lower than  $Q_{\alpha}$  (gs). If  $1/2^{-1}$ could be assigned to the 283.0 keV level is questionable, since a) a large energy increase of  $\approx 140$  keV would be required from  ${}^{205}$ Po to  ${}^{207}$ Rn and b) further  $3/2^{-}$  levels have been identified at energies of 595.11 keV and 384.34 keV in  $^{203}\mathrm{Pb}$  and  $^{205}\mathrm{Po},$  respectively. A  $3/2^-$  assignment would also be reasonable for this level. Estimation of the conversion coefficient that could be used for the determination of the multipolarity of the transition could not be performed, since the energy of the  $\alpha$ -particles feeding the 283 keV level is close to that of the 6624 keV transition of  $^{213}\mathrm{Ra},$  which was produced with a yield higher than two orders of magnitude. Therefore, the X-ray rate from the decays of the 283.0 keV level could not be determined.

#### 3.4 Isotope <sup>210</sup>Ra

The isotope <sup>210</sup>Ra has been first synthesized and identified by Valli *et al.* [12] using the reactions <sup>197</sup>Au(<sup>19</sup>F, 6n)<sup>210</sup>Ra and <sup>206</sup>Pb(<sup>12</sup>C, 8n)<sup>210</sup>Ra. They reported an  $\alpha$  energy and a half-life of (7018 ± 5) keV and (3.8 ± 0.2) s. The nuclear structure of the  $\alpha$ -decay daughter has been investigated by several groups either using in-beam  $\gamma$ -spectroscopy [15] or electron capture and positron decay of <sup>206</sup>Fr [17]. The first 2<sup>+</sup>-level is located at  $E^* = 575.3$  keV.

In our experiment we identified the  $\alpha$ -decay of <sup>210</sup>Ra into the first 2<sup>+</sup>-level of <sup>206</sup>Rn by coincidences between  $\gamma$ transitions of 575.3 keV and  $\alpha$ -particles of about 6451 keV. The result of our measurement is shown in fig. 2b. We obtained an  $\alpha$  energy of (6447 ± 5) keV and a relative intensity of  $\approx 0.0003$ . This result is remarkable since this value is an order of magnitude lower than the value of



**Fig. 6.** Suggested decay scheme for  $^{209}$ Ra. The energies are given in keV and refer to the  $Q_{\alpha}$ -values.

0.0037 expected from theoretical partial  $\alpha$  half-lives including spin hindrance [18, 19].

## 3.5 Isotope <sup>209</sup>Ra

The isotope <sup>209</sup>Ra has been first synthesized and identified by Valli *et al.* [12] using the reaction  $^{197}$ Au( $^{19}$ F,  $(7n)^{209}$ Ra. They reported an  $\alpha$  energy and a half-life of  $(7008 \pm 5)$  keV and  $(4.7 \pm 0.2)$  s. In our experiment we found two weak  $\gamma$ -lines of (633.7  $\pm$  1.1) keV and (387.0  $\pm$ 0.5) keV in coincidence with  $\alpha$ -particles of (6376 ± 10) keV and (6625  $\pm$  5) keV, respectively. The sums of the  $Q_{\alpha}$ values and the  $\gamma$  energies are close to the  $Q_{\alpha}$ -values for the known transitions of <sup>210,209</sup>Ra. We attribute the  $\gamma$ lines to  $^{205}$ Rn, since the energies do not fit with the decay of known levels in <sup>206</sup>Rn. A decay scheme is presented in fig. 6. Alpha-decay of the N = 119 isotone  $^{207}$ Rn feeds low-lying levels of <sup>203</sup>Po at  $E^* = 62.54$  keV (tentative spin and parity assignment:  $3/2^{-}$ ) and at 133 keV (tentative spin and parity assignment:  $1/2^{-}$ ) with relative intensities of 0.007 and 0.001, respectively. We did not observe coincidences between  $\gamma$  events below 200 keV and  $\alpha$ -decays in the range (6800–7000) keV, *i.e.* no  $\alpha$ -decay fine structure of <sup>209</sup>Ra similar to that of <sup>207</sup>Rn.

#### 4 Summary

Applying the technique of  $\alpha$ - $\gamma$  coincidence spectroscopy after in-flight separation of evaporation residues from the projectile beam, we succeeded to identify weak  $\alpha$  transitions of <sup>209-212</sup>Ra into excited levels of the daughter nuclei. For the even-even isotopes <sup>210,212</sup>Ra  $\alpha$ -decay into the

For the even-even isotopes  $^{210,212}$ Ra  $\alpha$ -decay into the known first 2<sup>+</sup>-levels of  $^{206,208}$ Rn was observed. For  $^{212}$ Ra the measured intensity agrees (within our experimental accuracy) with the expected value assuming an angular momentum change of  $\Delta L = 2$ , as it was also obtained for  $^{214}$ Ra [4]. For  $^{210}$ Ra, however, the observed intensity is an order of magnitude lower than expected.

For <sup>211</sup>Ra  $\alpha$ -decay into five excited daughter levels was observed. Unlike the neighbouring N = 121 isotone <sup>209</sup>Rn, no  $\alpha$ -decay into an expected low-lying  $1/2^{-1}$  state was observed. Therefore, the existence of such a state which is known at  $E^* = 143.166$  keV in  $^{205}$ Po and  $E^* = 126.51$  keV in  $^{203}$ Pb could not be proved in  $^{207}$ Rn in an energy range of (100–200) keV. Yet we observed  $\alpha$ -decay into the 665.0 keV level, known from the literature to be fed by  $\gamma$ -decay of the 0.18 ms  $13/2^+$  isomeric state at  $E^* = 899$  keV.

For <sup>209</sup>Ra only  $\alpha$ -decays into two excited levels of <sup>205</sup>Rn at  $E^* = 387$  and 634 keV were observed, but not into low-lying daughter levels at  $E^* < 200$  keV as is known for the N = 119 isotones <sup>207</sup>Rn and <sup>205</sup>Po.

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