

# The New Isotopes <sup>258</sup>105, <sup>257</sup>105, <sup>254</sup>Lr and <sup>253</sup>Lr

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Evaporation residues from the heavy-ion fusion reaction  ${}^{50}$ Ti on  ${}^{209}$ Bi were investigated. They were separated from the projectile beam by the velocity filter SHIP and identified after implantation into an array of position-sensitive surface-barrier detectors by analyzing their  $\alpha$ -decay chains. Spontaneous fission was also observed.

ing their  $\alpha$ -decay chains. Spontaneous fission was also observed. Four new  $\alpha$  emitters, <sup>258</sup>105 ( $T_{1/2} = 4.4^{+0.9}_{-0.6}$  s), <sup>257</sup>105 ( $T_{1/2} = 1.4^{+0.6}_{-0.3}$  s), <sup>254</sup>Lr ( $T_{1/2} = 13^{+3}_{-2}$  s), and <sup>253</sup>Lr ( $T_{1/2} = 1.3^{+0.6}_{-0.3}$  s) could be identified. For the isotope <sup>257</sup>105 we obtained a spontaneous-fission branch of about 20%. A spontaneous-fission activity with a halflife comparable to that for the  $\alpha$  decay of <sup>258</sup>105 was explained as fission of <sup>258</sup>104, formed by electron capture from <sup>258</sup>105.

An excitation function for evaporation-residue production was measured for bombarding energies in the range of  $E_{\rm CM}$ =184.4 MeV to  $E_{\rm CM}$ =196.6 MeV. Nearly all evaporation residues we observed, could be attributed to the 1n and 2n deexcitation channels. The maximum cross sections were  $\sigma(1n) = c/2.9 \pm 0.3$ ) nbarn, and  $\sigma(2n) = c/2.1 \pm 0.8$ ) nbarn, respectively.

We could measure the total kinetic energy of the fission fragments of  ${}^{258}104$  to be TKE=(220±15) MeV, a value that fits into empirical systematics based on a  $Z^2/A^{1/3}$  dependence.

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# 1. Introduction

The production of weakly excited compound nuclei in the region of the heaviest elements ( $Z \ge 100$ ) by complete fusion reactions using projectile – target combinations of isotopes close to the doubly magic <sup>48</sup>Ca and <sup>208</sup>Pb resp., was proposed by Oganessian et al. [1, 2] several years ago and proved unambiguously by observing the 1*n* deexcitation channel in the fusion reaction <sup>208</sup>Pb(<sup>50</sup>Ti, 1*n*)<sup>257</sup>104 [3, 4]. The importance of this discovery was shown clearly in attempts to produce elements with Z > 106. Although evidence was found, that, for the systems discussed in Refs. 1–4, complete fusion is hindered at bombarding energies close to the barrier, the method of cold fusion was the successful way to the new elements 107, 108, 109 [5–9].

In Refs. 3, 4, 6, 7, 9 the isotope identification was based on the observation of  $\alpha$ -decay chains, starting with the  $\alpha$  decay of the produced evaporation residue. It was shown, that an unknown isotope could be identified even by the observation of only one decay chain, if the decay properties of the daughter products are well known and sufficiently specific. For the unambiguous identification of the isotopes of element 107 and 109 reported in Refs. 6, 7 the most important links in the  $\alpha$ -decay chains are isotopes of element 105 with A < 260 and Lawrencium with A < 255.  $\alpha$ decay properties of these isotopes were completely unknown up to then. So it was the aim of our experiments to close this gap. Preliminary data of a first experiment on the synthesis of element 105 isotopes have been reported in Ref. 3. The most promising way to produce neutron-deficient isotopes of element 105 is the fusion of <sup>209</sup>Bi with <sup>50</sup>Ti [5]. According to our experiments on the synthesis of element 104 isotopes by fusion of <sup>208</sup>Pb and <sup>50</sup>Ti [4], the maximum of the evaporation-residue production is expected to lie about 3 MeV below the barrier calculated using a standard model [10, 11], despite of a possible hindrance of complete fusion in this range of bombarding energies.

#### 2. Experimental Method

## 2.1. Experimental Set-Up

The experiments were performed with a  ${}^{50}$ Ti beam of  $(0.5-1.8) \times 10^{12}$  particles per second from the UN-ILAC accelerator, Darmstadt, at specific projectile energies between (4.65–4.95) MeV/u. The energies were measured by a time-of-flight method monitoring the 27 MHz microbunches of the UNILAC beam [12] with an accuracy of  $\pm 0.02$  MeV/u. During the experiments the energy stability as well as the target quality

were controlled by monitoring the elastic scattering of the projectiles in the targets at  $30^{\circ}$  in combination with a time-of-flight method [13].

The experimental set-up used here, was identical to that used in the experiment to synthesize element 109 [9] and is described in detail in Refs. 14, 9. Therefore we will give only a brief summary in this paper.

The <sup>209</sup>Bi targets of (0.5-0.7) mg/cm<sup>2</sup> were covered on both sides with carbon layers each of 0.03 mg/cm<sup>2</sup> to improve radiative cooling [15], and were mounted on a rotating wheel [16].

The evaporation residues, which recoiled from the targets nearly unretarded, were separated from the projectile beam by the velocity filter SHIP [17], then passed a time-of-flight system, consisting of two large-area  $(30 \times 60 \text{ mm}^2)$  transmission detectors (TOF) [18] at a distance of 504 mm and were finally implanted into an array of seven position-sensitive surface barrier-detectors [14], where their kinetic energy as well as their subsequent  $\alpha$  decay or spontaneous fission was observed. The detectors were cooled to 260 K to obtain an energy resolution of 30 keV at FWHM and a position resolution of 0.3 mm at FWHM. In order to minimize the background in the energy region expected for the  $\alpha$  decays (7–10 MeV), which is due to scattered projectiles and target-like nuclei, passing SHIP with the same velocity as the evaporation residues, a plastic absorber foil of  $200 \,\mu g/cm^2$  was installed in front of the detector array. Behind the detector array, a high-purity germanium detector was mounted in close geometry to register y- or x-rays in coincidence to  $\alpha$  decays or spontaneous fission, respectively. A continuously running µs clock allowed to register the absolute time of each event observed in the detector array. This was later used to establish mother-daughter relationships by time correlations, and to determine halflives. The signals from the various detectors were registered and stored on tape event by event. A preliminary data analysis was done with a PDP11/45 computer using the GOLDA [19] program system, while a more thorough analysis was done off-line at the IBM 3081 system at GSI using the analysis system SATAN [19].

# 2.2. Calibrations

During the experiment the performance of SHIP was checked periodically by a set of test reactions  ${}^{50}\text{Ti} + {}^{107}\text{Ag}$ ,  ${}^{148}\text{Sm}$ ,  ${}^{170}\text{Er}$ , respectively. For the  $\alpha$ -energy calibration known  $\alpha$ -transition energies of isotopes produced in the test reactions were used. We took the recommended values of Ref. 20, if available, or the values published in Ref. 21. Since the nuclei were implanted into the detector the measured pulse height is the sum of the  $\alpha$  energy and a portion  $\Delta E_{\text{rec}}$  of

the recoil energy  $E_{\rm rec}$  of the  $\alpha$  daughter. The latter contribution was estimated to be  $\Delta E_{\rm rec}/E_{\rm rec} = 0.275$ , according to the results of Ref. 22.

The energy calibration of the recorded fission events was done in two steps: a) Extrapolation of the  $\alpha$ energy calibration from the energy region E <10 MeV to the region E = (100-250) MeV, which was done by use of a precision pulse generator, b) the estimation of the pulse-height defect (PHD) in the array detectors. For this purpose the test reactions were used. We took advantage of the fact, that in each reaction at least one known  $\alpha$  emitter was produced which had a halflife short enough to correlate the evaporation residues to  $\alpha$  particles. The absolute kinetic energy of these evaporation residues  $E_{abs}$ could be obtained from the time-of-flight and the known mass, whereas the apparent energy  $E_{app}$  was obtained from the pulse height of the array-detector signal. Energy losses in the upstream TOF  $\Delta E_{\text{TOF}}$  and in the dead layers of the array detectors  $\varDelta E_{dead}$  were calculated using data from published tables [23]. The pulse-height defect was then given by  $\Delta \varepsilon_{PHD} =$  $\varepsilon_{abs} - \varepsilon_{app} - \varDelta \varepsilon_{TOF} - \varDelta \varepsilon_{dead}$ .  $\varepsilon$  means the energy in LSS-units as defined by Lindhard et al. [24, 25]. The dependence of the logarithm of  $\Delta \varepsilon_{\rm PHD}$  on the logarithm of  $\varepsilon_{app}$  could be fitted by a third-order polynominal.

The energy calibration for the evaporation residues produced in the Ti+Bi bombardments, which was necessary for the mass determination of the heavy evaporation residues, was done in a similar way. Here we used in the test reactions the same absorbers in front of the detector array as in the irradiation of <sup>209</sup>Bi and did not correct the energy losses in the upstream TOF and the dead layer of the array detectors. The use of an effective PHD  $\Delta \varepsilon_{\text{PHD, eff}} = \varepsilon_{abs} - \varepsilon_{app}$  did not deteriorate the mass resolution, which was 15% FWHM.

Due to the mass criterion a sufficient discrimination between target-like nuclei and evaporation residues could be achieved, and an unambiguous assignment of  $\alpha$  decays and spontaneous-fission events, resp., to preceeding evaporation residues was achieved within  $\Delta t = 50$  s. The maximum time for  $\alpha$ - $\alpha$ -correlation of events within the beam pulse was limited to about 100 s, since the efficiency of the TOF, which was used as an anticoincidence, was only 90% for the background events in the energy range of the  $\alpha$  particles. For events within the beam pause  $\alpha$ - $\alpha$  correlation was possible up to 500 s.

#### 3. Experimental Results and Discussion

The <sup>209</sup>Bi targets were irradiated at four projectile energies. The projectile doses, the number of  $\alpha$  decays

Table 1. Summary of irradiation energies, projectile doses, numbers of observed  $\alpha$  decays and fission events attributed to ER

( <i>E</i> / <i>A</i> )/(MeV/u)	Projectiles $\times 10^{16}$	Σα	$\Sigma$ sf
4.65	0.6	0	1
4.75	16.2	82	47
4.85	1.69	6	4
(4.85	1.1	1	2) <sup>a</sup>
4.95	1.45	8	0
(4.95	0.7	1	1) <sup>a</sup>

\* In these experiments we had target problems. The results were not used to evaluate cross-sections

which were attributed to evaporation residues, and the number of observed fission events are listed in Table 1. The values in brackets refer to an experiment, where, due to target problems, formation cross sections could not be deduced.

The spectroscopic results are listed in Table 2. The errors for the halflives given in Table 2 as well as in the text are standard errors.

For the discussions of the  $\alpha$ -decay properties we use the concept of the 'hindrance factor (HF)'. In the literature [26-28] HF is often defined as the ratio  $HF = T_{\alpha}(exp)/T_{\alpha}(ee)$ .  $T_{\alpha}(exp)$  is the experimental  $\alpha$ halflife and  $T_{\alpha}(ee)$  is the corresponding partial halflife of an unhindered  $(\Delta I=0) \alpha$  transition.  $T_{\alpha}(ee)$  was calculated using the semi-empirical formula of Poenaru et al. [29] with the parameter modifications proposed by Rurarz [30]. It is known from literature (see e.g. Refs. 26-28), that for transuranium nuclei  $\alpha$  decay into daughter states, that correspond to the same Nilsson orbitals as the ground state of the mother, is often preferred, although those states may lie several hundred keV above the ground states. Those decays are called 'favoured transitions', their HF-values are often close to unity.

#### 3.1. $\alpha$ Emitters

In the bombardments of the  $^{209}$ Bi targets with  $^{50}$ Ti four new  $\alpha$  emitters were identified

a) a 4.4 s activity with four  $\alpha$  lines between 9.0 and 9.3 MeV was assigned to <sup>258</sup>105

b) a 1.4 s activity with three  $\alpha$  lines between 8.95 and 9.20 MeV was assigned to <sup>257</sup>105

c) a 13 s activity with two  $\alpha$  lines at 8.46 and 8.41 MeV was assigned to <sup>254</sup>Lr, the  $\alpha$  daughter of <sup>258</sup>105

d) a 1.3 s activity with two  $\alpha$  lines at 8.80 and 8.72 MeV was assigned to <sup>253</sup>Lr, the  $\alpha$  daughter of <sup>257</sup>105.

In addition to these activities some more  $\alpha$  emitters that could be assigned to later decay-chain members of <sup>258</sup>105 and <sup>257</sup>105 were observed. The identification of the new  $\alpha$  emitters was done by  $\alpha$ - $\alpha$  correla-

Isotope	$E_{lpha}/{ m keV}$	iα	Hindrance factor HF	$T_{1/2}/s$	$b_{lpha}$	$b_{\rm EC}$	$b_{ m sf}$
<sup>258</sup> 105	$9,299 \pm 15 \\9,172 \pm 15 \\9,078 \pm 15 \\9,008 \pm 15$	0.08 0.59 0.28 0.05	1,415 82 91 315	4.4 <sup>+0.9</sup> <sub>-0.6</sub>	0.67 <sup>+0.05</sup> <sub>-0.09</sub>	0.33 <sup>+0.09</sup> <sub>-0.05</sub>	-
<sup>257</sup> 105	$9,160 \pm 20$ $9,071 \pm 20$ $8,970 \pm 20$	0.3 0.3 0.4	37 20 7.5	$1.4^{+0.6}_{-0.3}$	$0.83 \pm 0.11$	_	$0.17 \pm 0.11$
<sup>254</sup> Lr	$8,460 \pm 20$ $8,408 \pm 20$	0.64 0.36	6.4 7.7	$13^{+3}_{-2}$	$0.78 \pm 0.22$	$0.22 \pm 0.06$	-
<sup>253</sup> Lr	$8,800 \pm 20$ $8,722 \pm 20$	0.56 0.44	6.7 4.9	$1.3^{+0.6}_{-0.3}$	>0.8	-	_
<sup>254</sup> No	$8,086 \pm 20$	1	2.0	$68^{+36}_{-18}$			
<sup>250</sup> Md	$7,837 \pm 20$ $7,751 \pm 20$	$\sim 0.2$ $\sim 0.8$	11.7 2.8	$40^{+37}_{-13}$	$0.13 \pm 0.10$	$0.87 \pm 0.10$	-
<sup>249</sup> Md	$8,026 \pm 20$	1	1.2	$25^{+14}_{-7}$	>0.6	-	-
<sup>245</sup> Es	$7,755\pm20$	1	2.5	$80^{+96}_{-28}$	$0.8^{+0.2}_{-0.5}$	$0.2^{+0.5}_{-0.2}$	-

Table 2. Summary of spectroscopic results

tions to known decay products

<sup>258</sup>105 –  $\alpha \rightarrow$ <sup>254</sup>Lr –  $\alpha \rightarrow$ <sup>250</sup>Md –  $\alpha \rightarrow$ <sup>246</sup>Es <sup>258</sup>105 –  $\alpha \rightarrow$ <sup>254</sup>Lr – EC  $\rightarrow$ <sup>254</sup>No –  $\alpha \rightarrow$ <sup>250</sup>Fm <sup>257</sup>105 –  $\alpha \rightarrow$ <sup>253</sup>Lr –  $\alpha \rightarrow$ <sup>249</sup>Md –  $\alpha \rightarrow$ <sup>245</sup>Es

where  ${}^{250}Md$ ,  ${}^{254}No$  and  ${}^{249}Md$  are known  $\alpha$  emitters.

It turned out, that  $^{258}105$  and  $^{257}105$  have  $\alpha$  lines with similar energies. If an  $\alpha$  decay was found to be correlated only to the evaporation residue and not to a daughter  $\alpha$  decay, no assignment to one of these two isotopes could be done.

As illustrative examples,  $\alpha$  spectra recorded at E/A =4.75 MeV/u are shown in Figs. 1a-1c. Figure 1a shows the  $\alpha$  spectrum recorded between the beam bursts, while Fig. 1b shows the spectrum of events anticoincident to the TOF following the implantation of a heavy nucleus with a recorded mass A > 230 within  $\Delta t = 22$  s at the position of implantation; accumulation of events with E = (8.9-9.4) MeV, that can be assigned to  $^{258}105$  and  $^{257}105$  and at  $E = (8.4 - 10^{10})^{10}$ 8.5) MeV, which is due to  $\alpha$  decays of <sup>254</sup>Lr, is seen. Figure 1 c shows the spectrum of  $\alpha$  particles following  $\alpha$  decays between E = (8.9-9.4) MeV within  $\Delta t = 500$  s at the same position. Two major groups at  $E = (8.05 - 10^{-1})^{-1}$ 8.15) MeV and E = (8.4-8.5) MeV are seen, which originate from <sup>254</sup>No and <sup>254</sup>Lr, both daughter products of <sup>258</sup>105, while a weaker group can be assigned to the granddaughter  $^{250}$ Md. Other  $\alpha$  events can be assigned to <sup>253</sup>Lr, <sup>249</sup>Md, <sup>245</sup>Es, which are decay products of <sup>257</sup>105.

Figure 2 finally shows the  $\alpha$  spectrum taken within 37 h after the end of the irradiation. Here one can

see the  $\alpha$  decays of the long living decay product <sup>246</sup>Cf of <sup>258</sup>105. Recent experiments, performed at Dubna, take advantage of the decay properties of this isotope, which was separated chemically from the target material and products from other reactions than complete fusion after the irradiation of <sup>209</sup>Bi with <sup>50</sup>Ti. Cross sections for the 1*n*-channel, deduced from the yield of <sup>246</sup>Cf  $\alpha$  decays are in good agreement to our results [31, 32].

# 3.1.1. The $\alpha$ -Decay Chain of <sup>258</sup>105

Isotope <sup>258</sup>105. This isotope was produced in the reaction <sup>209</sup>Bi (<sup>50</sup>Ti, *n*) <sup>258</sup>105 and identified by  $\alpha$ - $\alpha$  correlation to its daughter products <sup>254</sup>Lr, <sup>254</sup>No and <sup>250</sup>Md. Its production cross section peaked at  $E_{\rm CM} \simeq$ 189 MeV. The peak value was obtained to be  $\sigma_{\rm max} =$ (2.9±0.3) nb. The  $\alpha$  decays were divided into four groups with the energies  $E_{\alpha1...4} = 9,299, 9,172, 9,078,$ 9,008 keV. The halflife was measured to be  $T_{1/2} =$ (4.4 $^{+0.6}_{-0.6}$ ) s.

 $\alpha$  decay of this isotope is strongly hindered. For the most intensive  $\alpha$ -line we calculated a hindrance factor HF = 82, which is about one order of magnitude higher than that for the most intensive line of the neighbouring isotope <sup>257</sup>105. The HF values for the other transitions are considerably higher (see Table 2). Therefore one may conclude, that 'favoured transitions' were not observed. The reason may be, that the corresponding daughter levels lie more than 300 keV above the ground state and that  $\alpha$  decay to these states is strongly suppressed by the smaller *Q*-value. This behavior is similar to that of the odd-



Fig. 1.  $\alpha$ -Spectra observed in the irradiation of <sup>209</sup>Bi with <sup>50</sup>Ti at E/A = 4.75 MeV/u. a) Spectrum taken between the beam bursts (upper spectrum). b) Spectrum of  $\alpha$  decays following the implantation of an evaporation residue (measured mass > 230) within  $\Delta t = 22$  s. (medium spectrum). c) Spectrum of  $\alpha$  decays following  $\alpha$  decays of <sup>258</sup>105 or <sup>257</sup>105 within  $\Delta t = 500$  s (lower spectrum)

odd isotope <sup>252</sup>Es, which has also N=153 neutrons. Four x-ray events with an energy of  $E_x = (22.5 \pm 0.6)$  keV were observed in coincidence to  $\alpha$  decays with individual energies between  $E_{\alpha} = (9,056-9,129)$  keV, which were attributed to the line  $E_{\alpha3}$ . The measured x-ray energies are compatible with the theoretical energy  $E_{L\beta4} = 22.616$  keV [33] and the experimental value  $E_{L\beta4} = (22.61 \pm 0.18)$  keV published by Bemis [34] for the  $L\beta4$  line of Lawrencium. This indicates that the  $\alpha$  decay  $E_{\alpha3}$  populates a daughter state which decays to a high degree by  $L_{I}$  conversion. Therefore  $E_{\alpha3}$  is expected to be strongly influenced



Fig. 2.  $\alpha$ -Spectrum taken within 37 h after the end of the irradiation

by energy summing of  $\alpha$  particles and conversion electrons. Furthermore it can be assumed, that the transition probably is a magnetic one (*M*1), since for Lawrencium electric transitions should have a higher conversion coefficient for  $L_{II}$ -conversion than for  $L_{I}$ -conversion [35].

Isotope <sup>254</sup>Lr. This isotope was produced by  $\alpha$  decay of <sup>258</sup>105. The recorded  $\alpha$  events could be divided into two groups with energies  $E_{\alpha 1,2} = 8,460$ , 8,408 keV. The halflife is  $T_{1/2} = 13^{+3}_{-2}$  s. Some  $\alpha$  decays, that were attributed to <sup>258</sup>105, were found to be correlated to <sup>254</sup>No, indicating an EC-branch of <sup>254</sup>Lr. From the number of the observed correlations  $\alpha(^{258}105) \rightarrow \alpha(^{254}Lr)$  and  $\alpha(^{258}105) \rightarrow \alpha(^{254}No)$  we calculated, with the assumption that  $b_{\alpha}(^{254}No)=1$ , a value of  $b_{\rm EC} = 0.22 \pm 0.06$  for <sup>254</sup>Lr.

The HF-values for both transitions HF(8,460) = 6.4and HF(8,408) = 7.7 are almost equal and similar to those for the two most intensive  $\alpha$  transitions of <sup>258</sup>Lr. Thus the two observed lines may be regarded to belong to favoured transitions.

Isotope <sup>254</sup>No. This isotope was produced by electron capture of <sup>254</sup>Lr. The  $\alpha$  energy  $E_{\alpha} = (8,086 \pm 20)$  keV and the halflife  $T_{1/2} = (68^{+36}_{-18})$  s, as determined from our data are in good agreement to previously published values [36].

Isotope <sup>250</sup>Md. This isotope was produced by  $\alpha$  decay of <sup>254</sup>Lr. We determined the  $\alpha$ -decay energy, the halflife and, from the numbers of observed  $\alpha$  decays of <sup>254</sup>Lr and of <sup>250</sup>Md, the  $\alpha$  branching. Our experimental values  $E_{\alpha 1,\alpha 2} = 7,751, 7,837$  keV, with the relative intensities  $i_{\alpha 1} = 0.8$  and  $i_{\alpha 2} = 0.2, T_{1/2} = (40^{+37}_{-13})$  s and  $b_{\alpha} = 0.13 \pm 0.10$  are in good agreement to published literature values [36].

# 3.1.2. The $\alpha$ -Decay Chain of <sup>257</sup>105

Isotope <sup>257</sup>105. This isotope was produced in the reaction <sup>209</sup>Bi(<sup>50</sup>Ti, 2n)<sup>257</sup>105 and also identified by  $\alpha$ - $\alpha$ correlations to its decay products <sup>253</sup>Lr, <sup>249</sup>Md, <sup>245</sup>Es. The maximum production cross section of  $\sigma_{max} = (2.1 \pm 0.8)$  nb was observed at  $E_{CM} \simeq 197$  MeV. The  $\alpha$  decays were divided into three groups with energies  $E_{\alpha 1...3} = 9,160, 9,071, 8,970$  keV, which are similar to that of <sup>258</sup>105. The halflife was measured to be  $T_{1/2} = (1.4^{+0.6}_{-0.3})$  s.

For this isotope, the  $\alpha$  decay of  $E_{\alpha 3} = 8,970$  keV eventually may be regarded as the 'favoured transition' (see Table 2) because of its low HF value. It should be mentioned, however, that due to the small number of twelve observed counts, the relative intensities and thus the HF values are not very well established.

Isotope <sup>253</sup>Lr. This isotope was found in the  $\alpha$ -decay chains of <sup>257</sup>105. Two  $\alpha$  lines with mean energies  $E_{\alpha 1,2} = 8,800, 8,722$  keV could be attributed to it. The measured halflife is  $T_{1/2} = (1.3^{+0.6}_{-0.3})$  s.

 $\alpha$ -emission is the predominant decay mode of this isotope. From the numbers of recorded  $\alpha$  decays of  $^{257}105$  and  $^{253}$ Lr, resp., we estimate  $b_{\alpha} > 0.8$ .

The HF values for the observed transitions are HF (8,800) = 6.7 and HF (8,722) = 4.9. Similar values can be obtained for the most intensive  $\alpha$ -transitions of the neighbouring odd-even nuclei <sup>255</sup>Lr, <sup>257</sup>Lr and <sup>251</sup>Md. Thus one can conclude, that the observed transitions are favoured ones.

Isotopes <sup>249</sup>Md, <sup>245</sup>Es. These two isotopes were produced by  $\alpha$  decay of <sup>253</sup>Lr. Previously published values [36] for the energies and halflives could be reproduced within the error bars. Our experimental values are:  $E_{\alpha} = (8,026 \pm 20) \text{ keV}$ ,  $T_{1/2} = (25^{+14}_{-7})$  s for <sup>249</sup>Md and  $E_{\alpha} = (7,755 \pm 20) \text{ keV}$ ,  $T_{1/2} = (80^{+96}_{-28})$  s for <sup>245</sup>Es. From the observed  $\alpha$ -decay rates we calculated an  $\alpha$  branching  $b_{\alpha} > 0.6$  for <sup>249</sup>Md and  $b_{\alpha} = 0.8^{+0.2}_{-0.5}$ for <sup>245</sup>Es. Published values [37] are  $b_{\alpha} > 0.2$  for <sup>249</sup>Md and  $b_{\alpha} = 0.4 \pm 0.1$  for <sup>245</sup>Es. Taking into account our large standard errors due to the small number of only five observed counts, the agreement of our values to those of Ref. 37 is quite good.

We compared the  $\alpha$ -decay energies of the new  $\alpha$  emitters to the values of the known isotopes in a ' $Q_{\alpha}$ systematics' for isotopes with  $Z \ge 96$ . For the  $Q_{\alpha}$ value we took the highest known  $\alpha$ -transition energy. The data were taken from Ref. 38 (<sup>239</sup>Cf, <sup>243</sup>Fm, <sup>247</sup>Md), Ref. 4 (<sup>255</sup>104, <sup>256</sup>104), Ref. 39 (<sup>259</sup>106, <sup>260</sup>106, <sup>261</sup>106) and the data compilation of Ref. 36. Ground states of daughter nuclei may be populated only weakly by  $\alpha$  transitions. Therefore in some cases, where the  $\alpha$ -decay properties are only known scarcely



Fig. 3.  $Q_a$ -Systematics for the heaviest nuclei with  $Z \ge 96$ 

the ground-state  $\alpha$  transition may not have been observed up to now. The ' $Q_{\alpha}$  value' obtained here may be somewhat lower than the real Q value, which is the energy difference between the ground-state transitions. Fig. 3 shows that the ' $Q_{\alpha}$  values' for the new isotopes fit quite well into the general trend. It is furthermore seen, that the local minimum in the  $Q_{\alpha}$ values at N=152, which is associated with the deformed gap in the single-partical levels at this neutron number [40], is still observed for elements 103 through 105.

## 3.2. Spontaneous Fission

3.2.1. Assignment of the Spontaneous Fission Activities

Spontaneous-fission events were observed at all irradiation energies. Since an identification by motherdaughter correlations (like for the  $\alpha$  decay) is not possible here, the assignment of the spontaneous fission events is more difficult. We have based our identification on the observed production and decay properties in comparison to the identified  $\alpha$  activities. As a first step one can compare a) the production rates for  $\alpha$ -emitters and spontaneous fission at the different bombarding energies (see Table 1) and b) the halflives of the observed spontaneous-fission activities.

For the spontaneously fissioning events observed at 4.65 and 4.75 MeV/u, where  $\alpha$  decays from <sup>258</sup>105 (1*n*-deexcitation channel) are prevailing one gets  $T_{1/2}(\text{sf}) = (6.1^{+1.0}_{-0.8})$  s, which is compatible to the half-life of <sup>258</sup>105 (obtained from the  $\alpha$  decays), while for those observed at 4.85 and 4.95 MeV/u, where

 $\alpha$  decays from <sup>257</sup>105 (2*n*-deexcitation channel) are prevailing one gets  $T_{1/2}(\text{sf})=1.7^{+1.3}_{-0.5}$ ) s, which is, within the errorbars equal to the halflife of <sup>257</sup>105 obtained from the  $\alpha$  decays.

So, if we assign all spontaneous-fission events of the latter group to the 2n channel, we get an upper limit for the fission branch of  $^{257}105 b_{sf} = 0.17 \pm 0.11$ . With this fission branch for  $^{257}105$  and the number of  $\alpha$  decays of this isotope observed at E/A = 4.75 MeV/u, we expect less than one fission event of  $^{257}105$  at this bombarding energy. So we attribute all spontaneous fission events observed at E/A = 4.75 MeV/u to the 1n channel.

Since an electron-capture branch  $b_{\rm EC} = 0.27$  is expected for <sup>258</sup>105 from predictions of Kolesnikov and Demin [41], spontaneous fission of the EC-daughter  $^{258}104 (T_{1/2} = 11 \text{ ms} [42])$  is assumed to be the source of this activity. With the detector array we observe  $\alpha$  decay and spontaneous fission and do not register electron capture. The halflife for the spontaneousfission activity is calculated from the time differences between the implantation of the evaporation residues and the fission and thus is the combined halflife of <sup>258</sup>105 and <sup>258</sup>104. Since  $T_{1/2}$  (<sup>258</sup>105)  $\gg$   $T_{1/2}$  (<sup>258</sup>104) we get a value close to  $T_{1/2}$  (<sup>258</sup>105). An electron-capture branch  $b_{\rm EC} = 0.33^{+0.09}_{-0.05}$  for <sup>258</sup>105 was calculated from the number of observed  $\alpha$  decays and fission events, respectively. This interpretation of the fission activity seems more reliable than the assumption of spontaneous fission of the doubly odd isotope <sup>258</sup>105. From a comparison of the spontaneous fission halflives of <sup>255</sup>104, <sup>256</sup>104 [4], and <sup>257</sup>105, one can roughly estimate a hindrance per unpaired nucleon for spontaneous fission of the odd mass isotopes of a factor  $T_{\rm sf}(^{255}104)/T_{\rm sf}(^{256}104) \simeq 400$  and  $T_{\rm sf}(^{257}105)/T_{\rm sf}(^{256}104) \simeq 1,100$ , respectively. With the assumption that the influence of the unpaired nucleons will superimpose independantly, for the odd-odd isotope <sup>258</sup>105 a hindrance of a factor of  $10^{5}$ - $10^{6}$ , and thus a fission branch of  $b_{sf} \ll 0.01$  can be expected.

Finally we want to point to a striking difference of two standard errors between the halflife calculated from the  $\alpha$  decays  $(T_{1/2}^{\alpha} = (4.4_{-0.6}^{+0.9}) \text{ s})$  and from the fission events  $(T_{1/2}^{\text{sf}} = (6.1_{-0.8}^{+1.0}) \text{ s})$ . Thus an admixture of another fission activity cannot be excluded a priori. An analysis showed indeed, that the observed time distribution of the fission events is compatible with a  $(25\pm5)\%$  admixture of an activity with a halflive of  $T_{1/2} = (20\pm10)$  s. Due to the low excitation energy of  $E^* \simeq 17$  MeV such a possible activity could be assigned also to the 1*n*-channel. An isomeric state in  $^{258}105$  decaying by electron capture could be speculated. The observed fission activity would then again be due to  $^{258}104$ . One fission event, not regarded so far, was observed with a correlation time of 5 ms at an bombarding energy of 4.85 MeV/u. This time distance is hardly compatible with the halflives of the isotopes <sup>258</sup>105 and <sup>257</sup>105. We attribute it to a spontaneously fissioning doubly even isotope of element 104. From the two possible candidates, <sup>258</sup>104 ( $T_{1/2}=11$  ms [41]) and <sup>256</sup>104 ( $T_{1/2}=7.4$  ms [4]), the former is more likely due to the low excitation energy of the compound nucleus of  $E^* = (21.0^{+3.5}_{-1.5})$  MeV.

# 3.2.2. Kinetic Energy of the Fission Fragments of <sup>258</sup>104

We tried to obtain the total kinetic energy TKE of the fission fragments of <sup>258</sup>104. Since the range of the fission fragments is larger than the implantation depth of the evaporation residues, in general, the recorded energy signal from a spontaneous-fission event is the sum of the kinetic energy of one fission fragment and the energy loss within the detector of the other fragment, and therefore lower than TKE.

Having this in mind, we tried to reproduce the measured spectrum with a Monte-Carlo calculation. We took into account the pulse-height defect as described in Sect. 2.2, the implantation depth, the range straggling, TKE and its standard deviation  $\sigma_{TKE}$ , the mass split and its standard deviation  $\sigma_A$ , as well as neutron emission from the fragments. Details of this procedure are given in Refs. 4, 43.

We obtained a mean value of TKE =  $(220 \pm 15)$  MeV from the Monte-Carlo calculations, a value that agrees rather well with results expected from empirical systematics of Unik et al. [44] (215 MeV) and Viola et al. [45] (209 MeV), which are based on a  $(Z^2/A^{1/3})$ dependence of the fission energies. The result of the calculations is shown in Fig. 4. Despite the limited accuracy of the measured TKE, it can be concluded that the fission of <sup>258</sup>104 does not belong to the cases discussed in Ref. 46, which were found to have TKE values, which are about 40 MeV higher than expected from the systematics. Our measurement does not allow us to distinguish between symmetric and asymmetric fission however.

#### 3.3. Excitation Functions

The irradiations at specific beam energies of (4.65-4.95) MeV/u showed, that 1n- and 2n-deexcitation channels are dominating in this energy region, the cross sections are listed in Table 3. The indicated errors are statistical errors only. An excitation function for evaporation residue production is shown in Fig. 5. It is characterized by a sharp decrease for  $E_{\rm CM} < 188$  MeV and a rather flat dependence in the



Fig. 4. Comparison between the measured energy distribution (full line) of registered spontaneous fission events of  $^{258}104$  and a Monte-Carlo-calculation for TKE = 220 MeV (dotted line)

**Table 3.** Summary of measured evaporation residue and xn cross sections

$\sigma_{ m ER}/{ m nb}$	$\sigma_{1n}/nb$	$\sigma_{2n}/nb$
$2.7 \pm 0.8$	$0.6 \pm 0.3$	$2.1 \pm 0.8$
$2.6 \pm 0.3$		_
$3.2 \pm 0.3$	$2.9 \pm 0.3$	$0.3^{+0.3}_{-0.15}$
$0.5 \pm 0.3$	$0.5 \pm 0.3$	_
	$\sigma_{\rm ER}/\rm nb$ 2.7±0.8 2.6±0.3 3.2±0.3 0.5±0.3	$\sigma_{ER}/nb$ $\sigma_{1 n}/nb$ 2.7 ± 0.8         0.6 ± 0.3           2.6 ± 0.3         -           3.2 ± 0.3         2.9 ± 0.3           0.5 ± 0.3         0.5 ± 0.3



Fig. 5. Measured excitation function for the production of evaporation residues for the system  $^{50}\text{Ti} + ^{209}\text{Bi}$ . (The line is to guide the eyes)

region  $E_{CM} = (188-197)$  MeV. The Bass-barrier for this system is  $E_B = 196$  MeV [10]. Evaporation residue production was found to occur even 8 MeV below  $E_B$ , a situation, similar to the system  ${}^{50}\text{Ti} + {}^{208}\text{Pb}$ [4]. In Ref. 4 it was concluded, that complete fusion is hindered by a factor of about 30 at bombarding energies close to the classical fusion barrier for such heavy systems. But  $E_B$  still seems to be a good guess for choosing bombarding energies to produce heavy isotopes with target-projectile combinations similar to that used here.

The search for other open deexcitation channels showed only a positive result for a small *p*-channel of  $\sigma_p = 0.1^{+0.1}_{-0.05}$  nb at  $E^* = 21$  MeV.  $\alpha$ -decays or spontaneous fission events that could be attributed to other channels ( $\gamma$ , 3n, pn, p2n, 2p, 2pn and channels involving the emission of  $\alpha$ -particles) unamabiguously were not observed. Upper limits for these channels are  $\sigma \leq (0.05-0.1)$  nb at  $E^* = 16.5$  MeV and  $\sigma \leq (0.3-0.6)$  nb at  $E^* = 21$  MeV and  $E^* = 25$  MeV, respectively.

#### 4. Conclusion

In our experiments four new isotopes  $^{253}$ Lr,  $^{254}$ Lr,  $^{257}105$ ,  $^{258}105$  could be identified unambiguously by their  $\alpha$ -decay properties. The existance of a 5 s-spontaneous-fission activity observed in the reaction  $^{50}$ Ti +  $^{209}$ Bi and assigned to the isotope  $^{257}105$  in Ref. 5, could be confirmed. Our analysis showed, that it consists of at least three components, SF of  $^{257}105$  (2*n*-channel), SF of  $^{258}104$ , produced by EC-decay of  $^{258}105$  (1*n*-channel), and SF of  $^{258}104$  (1*p*-channel).

From the energy distribution of the registered fission events, we estimated the total kinetic energy release in spontaneous fission of <sup>258</sup>104. Our value is, within our limited accuracy, in good agreement with the predictions from empirical systematics [44, 45].

An analysis of the production rates showed, that the evaporation residues originate predominantly from the 1n- and 2n-deexcitation channels of the compound nucleus  $^{259}105$  in the region of bombarding energies considered here, i.e. close to the classical fusion barrier. The cross sections for other deexcitation channels were found to be at least about one order of magnitude lower.

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