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EXPERIMENTS ON THE SYNTHESIS OF NEW SUPERHEAVY ELEMENTS*

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Evaporation residue production in cold fusion reactions of Pb- and Bi- target nuclei with projectiles of elements between Ti ($Z = 22$) and Zn ($Z = 30$) leading to compound nuclei $Z_{CN} = 104 - 113$ was investigated at SHIP. The result was the first unambiguous identification of isotopes of elements $Z = 110$, $Z = 111$ and $Z = 112$ in bombardments of ^{208}Pb ,^{209}Bi with ^{62,64}Ni, ^{70}Zn. We further report on the identification of ^{256}Db, ^{252}Lr and on new, improved decay data of ^{253}Lr, ^{255}Rf, ^{257}Db, allowing a first deeper insight in the nuclear structure of these isotopes.

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1 Introduction

More than thirty years ago early extensions of the nuclear shell model to regions of nuclei having proton and neutron numbers considerably higher than the heaviest ones known at that time led to the prediction of closed spherical nuclear shells at $Z = 114$ and $N = 184$ [1]. Nuclei close to the crossing point of both shells were predicted to be stabilized against spontaneous fission by large shell correction energies and consequently long half-lives up to values comparable to the age of the universe were expected. Much effort to produce and to detect these 'superheavy' nuclei in the laboratory as well as to search for them in nature has been brought up since these days - however, unsuccessfully up to now. Nevertheless theoretical models have been refined considerably and experimental work has produced heavier and heavier nuclei, slowly approaching the magic number of $Z = 114$. The present situation is illustrated in Fig. 1, where ground-state shell correction energies obtained by Smolanczuk et al. [2] using a Nilsson-Strutinsky approach are plotted as function of N and Z. Two regions of enhanced nuclear stability are visible: one around $Z = 114$ and $N = 184$, representing the 'classical' superheavy element region and a second one around $Z = 108$ and $N = 162$ with almost equal maximum shell correction energies but β_2 - values of up to $\approx$0.24, representing 'deformed' nuclear shells.

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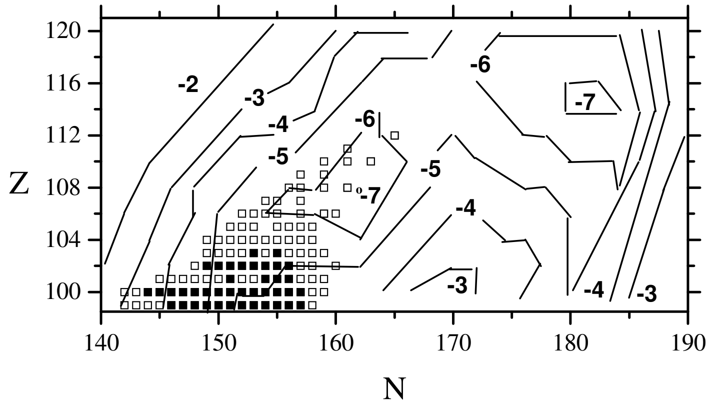


Fig. 1. Ground state shell correction energies according to [2]. The squares represent the presently known nuclei, the full squares those already known at early days of superheavy element predictions (ca. 1966).

The search for superheavy elements was a substantial motivation for the construction of the UNILAC and the velocity filter SHIP [3] at GSI in Darmstadt, and research work in this field has been carried out since about twenty years at these facilities. In the present paper we will report on the results of a series of experiments performed in the period 1994 - 1998 aiming at evaporation residue production in reactions leading to compound nuclei with atomic numbers $Z = 104 - 113$. For details on the experimental set-up we refer to [4].

2 Synthesis of new elements $Z > 109$

2.1 Excitation functions

Although 'cold' fusion of lead or bismuth targets with medium heavy projectiles like ^{50}Ti or ^{64}Ni has proven to be up to now the most successful method for the production of transactinide nuclei, we are faced with the fact that maximum cross sections typically decrease by a factor of ≈ 3.5 when the atomic number of the compound nucleus is increased by one (Fig. 2). Since, on the other hand, the formation cross section of a specific nuclide in a given reaction is strongly dependent on the excitation energy E^* of the compound nucleus the choice of the optimum value is crucial for the successful production of heaviest nuclei. We decided to determine it empirically by extrapolating the results from measured excitation functions.

Excitation functions, recently measured at SHIP for the systems ^{50}Ti, ^{58}Fe, and ^{64}Ni + ^{208}Pb are shown in Fig. 3a-c. Excitation energies, referring to the center of the target, were calculated using experimental mass excesses published by Wapstra and Audi [5] and values predicted by Myers and Swiatecki [6]. The data in Fig. 3 indicate a clear trend of decreasing excitation energy for the maxima of the 1n de-excitation channel with increasing atomic number. While for ^{50}Ti + ^{208}Pb the maximum for the 1n - cross section was obtained at $E^* = (15.6 \pm 0.1)$ MeV (Fig. 3a) it decreased to $E^* = (13.6 \pm 0.2)$ MeV for ^{58}Fe + ^{208}Pb (Fig. 3b). The widths of the 1n - de-excitation functions are narrow, values of (4-5) MeV (FWHM) were obtained from a Gaussian fit to the data.

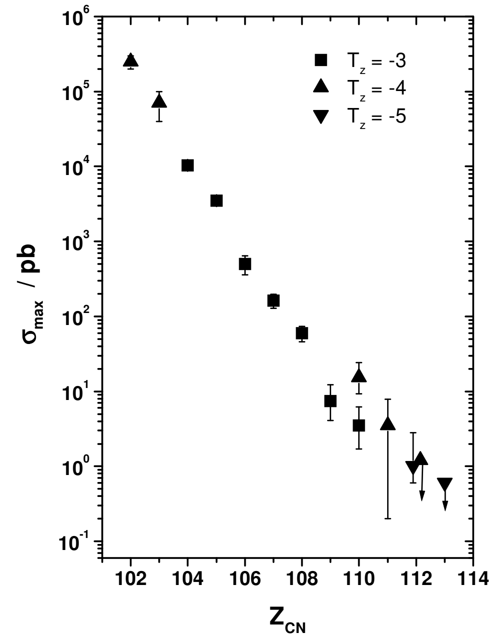


Fig. 2. Systematics of 1n- cross-sections for heavy element production in cold fusion reactions. T_z denotes the isospin of the projectile.

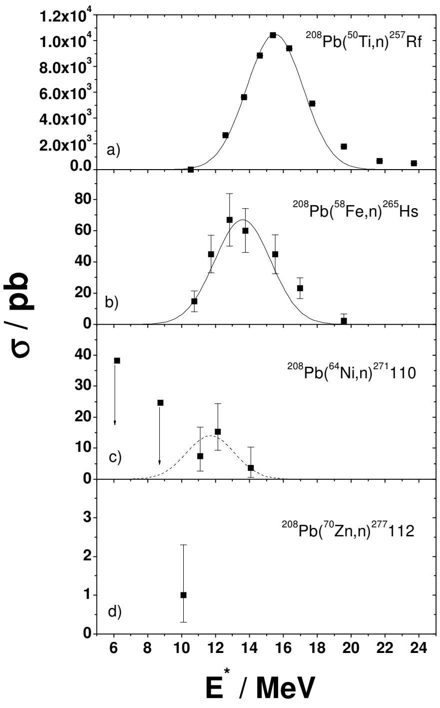


Fig. 3. Excitation functions for ^{50}Ti + ^{208}Pb (a), ^{58}Fe + ^{208}Pb (b), ^{64}Ni + ^{208}Pb (c), ^{70}Zn + ^{208}Pb (d).

2.2 Element 110

On the basis of these results an optimum value of $E^* = 12.3$ MeV for the production of $^{269}110$ by ^{208}Pb(^{62}Ni,1n)^{269}110 was extrapolated. In an experimental run of 12 days (total projectile dose: 2.2 x 10^{18}) we observed four α- decay chains starting with a decay of $^{269}110$. A mean energy $E_\alpha = 11.112 \pm 0.02$ MeV and a half-life of $T_{1/2} = 170^{+160}_{-70}\mu$s were obtained. The formation cross section was $\sigma = 3.5^{+2.7}_{-1.8}$ pb.

Since for the more neutron rich projectile ^{64}Ni a higher cross section could be expected, allowing to check the validity of our extrapolation, in a succeeding experiment a rough excitation function was measured (Fig. 3c). From fitting a Gaussian to the data the maximum of the 1n-channel was settled at $E^* = (12.4 \pm 0.5)$ MeV. Altogether nine α - decay chains that were attributed to $^{271}110$ were observed. They were subdivided into three groups: five events decay with a mean energy $E_\alpha = 10.738$ MeV, two events with $E_\alpha = 10.681$ MeV. The time differences between the implantation of the evaporation residues and the succeeding α-decays $\Delta t(ER-\alpha)$ lay between 0.2 ms and 4.4 ms, resulting in a half-life $T_{1/2} = 1.1^{+0.6}_{-0.3}$ ms. The third group existed of only one decay chain starting with an energy of $E_\alpha = 10.709$ MeV, having $\Delta t(ER-\alpha) = 81$ ms. Because of this significantly longer lifetime, it seems justified to assign this transition to a second level of ^{271}110.

2.3 Element 111

The positive results on the synthesis of element 110 encouraged us to immediately start an attempt to produce an isotope of element 111 by the reaction ^{209}Bi(^{64}Ni,1n)^{272}111. Since the extrapolation of the optimum projectile energy, derived from the doubly even nucleus ^{208}Pb, seemed less certain for the odd-even nucleus ^{209}Bi at that time, we irradiated at three different projectile energies E^* = 10.0, 11.6 and 13.0 MeV, starting from the lowest value that seemed meaningful. While no event that could be attributed to ^{272}111 was observed at $E^* = 10.0$ MeV ($\sigma < 2.9$ pb (68 % confidence limit)), one event was registered at $E^* = 11.6$ MeV ($\sigma = 1.7^{+3.3}_{-1.4}$ pb), and two more at $E^* = 13.0$ MeV ($\sigma = 3.5^{+4.6}_{-2.3}$ pb). α- energy and half-life are $E_\alpha = 10.82 \pm 0.02$ MeV and $T_{1/2} = 1.5^{+2.0}_{-0.5}$ ms.

2.4 Element 112

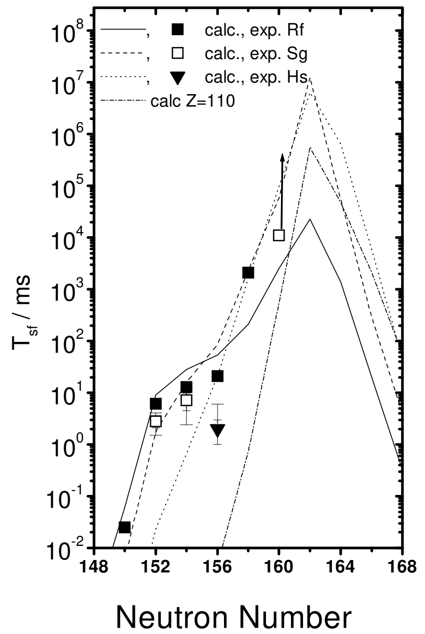
On the basis of the previous results an optimum value $E^* = 10.1$ MeV was chosen for production of ^{277}112 by the reaction ^{208}Pb(^{70}Zn,1n)^{277}112. In an experimental run of about six weeks, where a total projectile dose of 3.4 x 10^{18} was collected, two α-decay - chains that were attributed ^{277}112 were observed. They started with decays of $E_\alpha = 11.65$ MeV and $E_\alpha = 11.45$ MeV; the half-life obtained is $T_{1/2} = 240^{+430}_{-90} \mu$s. The resulting production cross section is $\sigma = 1.0^{+1.0}_{-0.4}$ pb (Fig. 3d).

2.5 Attempt to produce element 113

A search for the next heavier element with $Z = 113$ was performed in spring 1998. On the basis of the data for lighter elements a cross section between 0.3 pb and 1 pb was expected. During an irradiation period of 46 days, using the ^{70}Zn+^{209}Bi reaction at two slightly different beam energies of $4.97 \cdot A$ MeV (4.5×10^{18} projectiles) and $5.00 \cdot A$ MeV (3.0×10^{18} projectiles) no event was observed that could be assigned to a decay chain starting at element 113. The obtained cross section limits are $\sigma < 0.9$ pb and $\sigma < 1.4$ pb, respectively, resulting in an averaged value of $\sigma < 0.6$ pb at a probability level of 68 %.

3 Enhanced nuclear stability close to $N = 162$

As a consequence of the local maximum of ground state shell correction energies at $N = 162$ and $Z = 108$ (see Fig. 1) an enhanced stability of nuclei against spontaneous fission, and to lower extend also against α- decay is expected in this region [7]. In Fig. 4 we have displayed calculated [7] and experimental fission halflives for even - even isotopes $Z \geq 104$. Qualitatively the experimental data indicate a strong increase in T_{sf} approaching $N = 162$, thus supporting the enhanced stability predicted by theory. However no data are available for $N \geq 162$ so the maximum in T_{sf} is not localized. From this point we may conclude that the experimental data express evidence for a neutron shell but do not allow to position it.



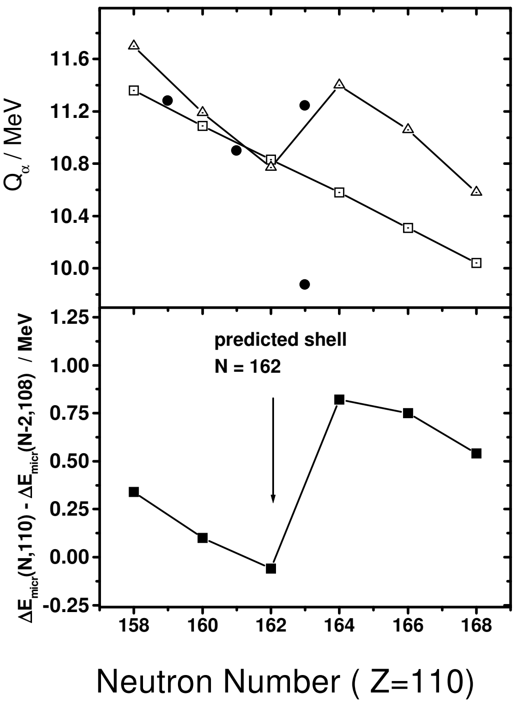


Fig. 4. Comparison of calculated [7] and experimental fission halflives for even-even nuclei $Z = 104$ to $Z = 110$.

Fig. 5. Experimental and calculated Q_α-values for $Z = 110$ isotopes. • $Q_{\alpha,exp}$, □ $Q_{\alpha,LD}$[6], △ $Q_{\alpha,LD+shellcorr.}$[2].

Experimental evidence for a neutron shell also can be obtained from Q_α-systematics for an isotopic chain. A shell crossing is indicated by a local minimum in the Q_α-values. This is demonstrated in Fig. 5. The open squares in the upper part represent calculated Q_α-values from liquid drop masses, which results in a straight line, decreasing with increasing neutron number. Introducing shell effects, a structure with a local minimum at the predicted shell at $N = 162$ (open triangles) becomes evident. It is effected by the differences of the ground state shell correction energies of the mother and daughter nuclei, as shown in the lower part of Fig. 5.

In the case of α - decay of $^{277}112$ the predicted shell at $N = 162$ is crossed by $^{273}110_{163} \stackrel{\alpha}{\rightarrow}$ $^{269}Hs_{161}$. In the two observed chains for $^{277}112$ [8] striking different α- energies and 'lifetimes' (i.e. $\Delta t(\alpha(^{277}112) - \alpha(^{273}110)))$ of $E_\alpha = 9.73$ MeV, $\Delta t(\alpha(^{277}112) - \alpha(^{273}110)) = 170$ ms and $E_\alpha = 11.08$ MeV, $\Delta t(\alpha(^{277}112) - \alpha(^{273}110)) = 110$ μs were measured. Because of the large difference in the time distances the two transitions must be assigned to different levels in $^{273}110$. Calculated life-times result in values of 175 ms and 67 μs, respectively, both for $\Delta I = 0$ transitions, which compare well with the measured time distances. We conclude that both transitions are unhindered and connect analogous states in mother and daughter nuclei. It is evident in Fig. 5 (full dots) that the higher energy causes a local minimum, indicating a shell crossing, while the lower energy indicates a continuation of the trend of decreasing Q_α- values with increasing

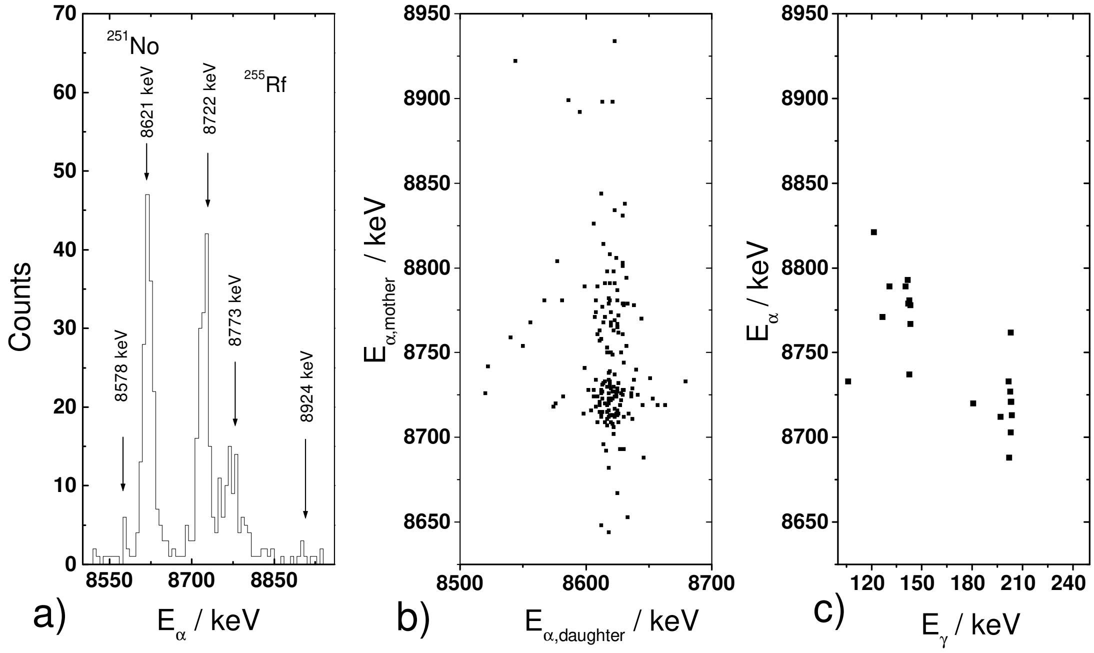


Fig. 6. Experimental results for ^{255}Rf; a) α- spectrum of correlated events; b) α - α correlation plot for ^{255}Rf - ^{251}No; c)observed α - γ coincidences for ^{255}Rf.

neutron numbers. Since it is not clear presently which energy has to be attributed to the ground state decay and which to the decay of an isomeric state, we cannot regard our results as a proof (or a disproof) for a neutron shell at $N = 162$. More data are necessary. It certainly would be helpful to produce the even-even isotopes ^{268,270,272}110 by irradiation of ^{207}Pb with ^{62,64}Ni and ^{70}Zn, since ground state to ground state transitions are expected to be the most intense ones in even - even nuclei.

4 Decay properties of the neutron deficient nuclei $Z = 103 - 105$

The improved sensitivity of our set-up does not only allow us to search for new elements on the pb - level within reasonable irradiation times (in the order of some weeks), but also allows for the production of a 'large' (>100) number of nuclei for isotopes having production cross sections of typically (0.1-10) nb. Thus a more detailed investigation of the α- and sf properties and also α-γ - coincidence measurements become possible. First detailed results on the α - fine structure had been obtained for ^{257}Rf, [9]. Here we will report on more recent results on ^{255}Rf, ^{257,256}Db, ^{253,252}Lr.

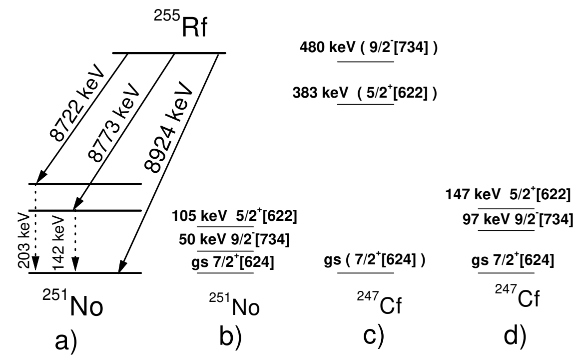


Fig. 7. a) tentative decay scheme for ^{255}Rf; b), d) calculated level schemes for ^{251}No, ^{247}Cf [10], c) level scheme attributed to ^{247}Cf (simplified) [11].

4.1 ^{255}Rf

In a previous study of evaporation residues from ^{50}Ti + ^{206,208}Pb, an indication for the existence of a low lying isomeric state in ^{255}Rf decaying by α- emission with $T_{1/2} \approx 0.9$ s seemed evident, since an abnormally low number of α- decays in the interval $E_\alpha = (8720-8730)$ keV was followed by daughter decays (^{251}No) [9]. However, neither from theoretical predictions [10] nor from the known levels of lighter $N = 151$ nuclei with even Z numbers (^{251}Fm, ^{249}Cf, ^{247}Cm, ^{245}Pu) [11] having a similar structure, a level sequence allowing the existence of an isomeric state of $T_{1/2} \approx 1$ s could be expected. To clarify this problem we used the ^{207}Pb(^{50}Ti,2n)^{255}Rf reaction to produce an order of magnitude more events of ^{255}Rf. The result is shown in Fig. 6a,b. On the basis of a considerably higher number of observed decays we were not able to confirm an 'abnormal low' correlation rate for $E_\alpha = (8720-8730)$ keV within the statistical uncertainty.

In addition we observed a few γ - events in coincidence to the α- decays of ^{255}Rf. Two different groups are indicated (Fig. 6c), a) γ-events with a mean energy of $E_\gamma = 203 \pm 3$ keV essentially in coincidence to α- decays of $E_\alpha = 8722 \pm 10$ keV, and b) $E_\gamma = 142 \pm 3$ keV coincident to $E_\alpha = 8773 \pm 10$ keV. The sums $E_\alpha + E_\gamma = 8925$ keV and $E_\alpha + E_\gamma = 8915$ keV are equal within 10 keV. They are also close to the mean energy $E_\alpha = (8924 \pm 10)$ keV of the group of α-decays in the interval $E_\alpha = (8908-8936)$ keV being the α-decays with the highest energies followed by decays of ^{251}No. So we conclude that both γ- transitions lead to the ground state of ^{251}No (Fig. 7a); a calculated level scheme [10] is shown in Fig. 7b. Due to the lowest hindrance factor of HF≈4 the $E_\alpha = 8722$ keV transition is assigned to the 'favored' transition, leading to the analogous Nilsson state. Striking, however, is the low value HF≈13 for the $E_\alpha = 8773$ keV transition. In the lighter $N = 149$ isotones the first excited Nilsson level is settled as 5/2$^+$[622] [11]. The relative intensities of decays into this level are <0.05 in these nuclei. Assigning the $E_\alpha = 8773$ keV - line to the transition 9/2$^-$[734] → 5/2$^+$[622] would result in a striking high relative intensity $i_{\alpha,rel} \approx 0.25$ and hence an abnormal low hindrance factor. In addition, if the γ - decay indeed leads to the ground state in ^{251}No, the 9/2$^-$[734] Nilsson level would lie considerably lower than in the lighter $N = 149$ nuclei.

Thus both observations may indicate a change in the nuclear structure for $N = 149$ isotones be-

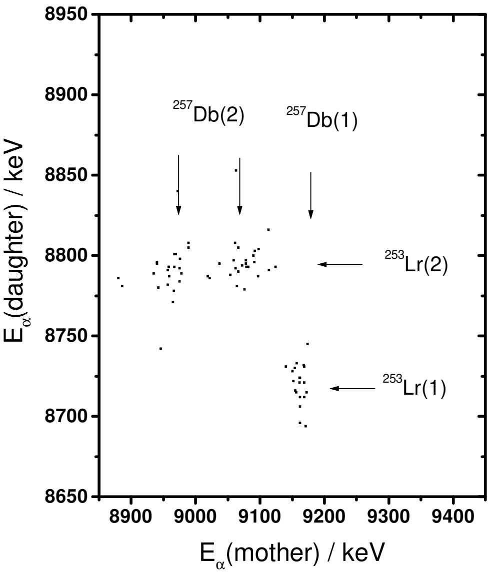


Fig. 8. α - α correlation plot for ^{257}Db - ^{253}Lr.

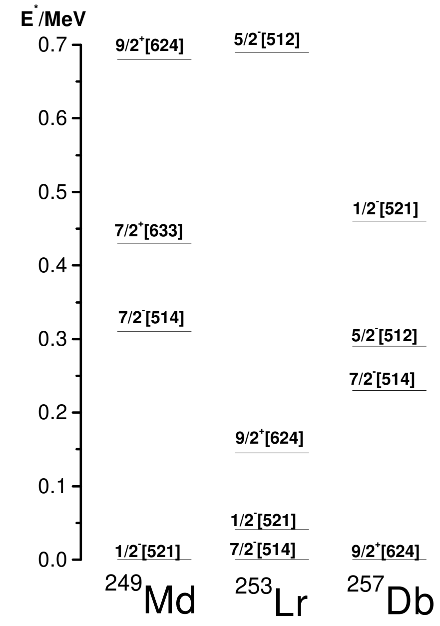


Fig. 9. Theoretical level schemes ^{257}Db, ^{253}Lr, ^{249}Md [10].

tween $Z = 98$ and $Z = 102$. On the basis of the present data any definite conclusions would be too speculative. More experimental data are necessary.

4.2 ^{257}Db, ^{253}Lr

In our first irraditions of ^{209}Bi with ^{50}Ti [12] ^{257}Db and ^{253}Lr were identified by delayed $\alpha - \alpha$ - coincidences to their decay products ^{249}Md and ^{245}Es. Three α-lines of E_α = 9160, 9071, 8970 keV were attributed to ^{257}Db, and two lines of E_α = 8800, 8722 keV were assigned to ^{253}Lr. A total of 12 α- decays of ^{257}Db correlated to any daughter product ^{253}Lr, ^{249}Md or ^{245}Es had been observed then.

In a recent experiment, the evaporation residue production for the reaction ^{50}Ti + ^{209}Bi was investigated in a bombarding energy range of $E = 4.59 \cdot A$ MeV to $5.08 \cdot A$ MeV. We observed about 200 α-decays of ^{257}Db correlated to any of the daughter products. The previously reported α-energies of ^{257}Db and ^{253}Lr could be reproduced within the error bars (see Table I). With respect to the correlations, however, a behavior was found, that was not realized in the previous experiment due to the small number of observed events: α-decays of ^{257}Db of E_α = 9163 keV were only followed by α-decays of ^{253}Lr of E_α = 8723 keV, while those ^{257}Db decays of E_α = 9074, 8967 keV were only followed by ^{253}Lr decays of E_α = 8794 keV (see Fig. 8). No statistically significant difference in the α-decay energies of the granddaughter ^{249}Md was found. Calculated level schemes [10] at low excitation energies are shown for ^{257}Db, ^{253}Lr, ^{249}Md. A possible candidate for an isomeric state is the $1/2^-[521]$ level. Weisskopf estimates indicate halflives in the order of some tenth of a second for $1/2^- \rightarrow 7/2^-$ transitions for energy differences $\Delta E \approx 100$ keV or lower. In the case of ^{257}Db this means that the $5/2^-$ level should lie above the $1/2^-$. However, definite conclusions on the level structure cannot be drawn on the basis of the present data, thorough α-γ- and α - conversion electrons coincidence measurements are necessary.

Estimation of an sf-branch of ^{257}Db was rendered more difficult by the fact, that as well for the 1n - channel as for the 3n - channel sf is observed via the decay path $^{258}\mathrm{Db} \overset{EC}{\rightarrow} {}^{258}\mathrm{Rf} \overset{sf}{\rightarrow}$ (1n) or $^{256}\mathrm{Db} \overset{EC}{\rightarrow} {}^{256}\mathrm{Rf} \overset{sf}{\rightarrow}$ (3n). The search for sf of ^{257}Db was therefore restricted to bombarding energies where contributions σ(1n) and σ(3n) were less than ten per cent of that of the 2n - channel. An upper limit of three sf events that could be attributed to ^{257}Db(1) or ^{257}Db(2) were registered, resulting in upper limits of $b_{sf} < 0.13$ for ^{257}Db(1) and $b_{sf} < 0.06$ for ^{257}Db(2). Furthermore three α-sf correlations were observed. According to E_α two sf events were attributed to ^{253}Lr(1) and one to ^{253}Lr(2). Resulting branchings are: $b_{sf} = 0.05 \pm 0.04$ for ^{253}Lr(1) and $b_{sf} = 0.01^{+0.01}_{-0.005}$ for ^{253}Lr(2).

4.3 ^{256}Db, ^{252}Lr

Table I. Summary of decay properties of ^{251}No, ^{252,253}Lr, ^{255}Rf, ^{256,257}Db, * = only one count observed

Isotope	$E_{\alpha 1}$ (keV)	i_{rel}	$T_{1/2}$ (s)	
^{251}No	8621±10	$0.96^{+0.02}_{-0.06}$	0.76±0.03	$b_{sf} < 0.0026$
	8578±15	0.04±0.015		
^{252}Lr	9018±20	0.73±0.26	$0.36^{+0.11}_{-0.07}$	
	8974±20	0.27±0.07		
^{253}Lr(1)	8723±10		$1.49^{+0.30}_{-0.21}$	b_{sf} = 0.05±0.04
^{253}Lr(2)	8794±10		$0.57^{+0.07}_{-0.06}$	$b_{sf} = 0.01^{+0.01}_{-0.05}$
^{255}Rf	8722±10	0.55±0.05	1.64±0.11	b_{sf} = 0.45±0.06
	8773±10	0.25±0.03		
	8797±15	0.07±0.02		
	8692±15	0.035±0.010		
	8831±15	0.035±0.010		
	8887±15	0.025±0.011		
	8924±15	0.025±0.0113		
^{256}Db	9018±20	≈0.64	$1.5^{+0.5}_{-0.4}$	b_{EC} = 0.35±0.12
	8891±20*	≈0.12		
	9075±20*	≈0.12		
	9120±20*	≈0.12		
^{257}Db(1)	9163±10		$0.76^{+0.15}_{-0.11}$	b_{sf} <0.13
^{257}Db(2)	9074±10	0.38±0.08	$1.50^{+0.19}_{-0.15}$	b_{sf} < 0.06
	8967±10	0.33±0.08		
	8937±15	0.09±0.04		
	9021±15	0.09±0.04		
	8892±15	0.055±0.031		
	9117±15	0.055±0.031		

Previously unknown α - activities were observed in the ^{50}Ti + ^{209}Bi reactions at the highest irradiation energies $E = 4.97 \cdot A$ MeV and $E = 5.08 \cdot A$ MeV. By delayed α-α - coincidences to known decay products ^{248}Md, ^{248}Fm and ^{244}Cf they were attributed to ^{256}Db and ^{252}Lr. Decay properties are listed in table 1.

In addition to the α- decays also nine sf events were registered at these irradiation energies and attributed to the 3n de-excitation channel. An estimation of T_{sf} for the odd-odd nucleus ^{256}Db on the basis of measured or calculated sf half-lives for the neighbouring even - even nuclei ^{256,258}Sg, ^{254,256}Rf and the sf hindrance factors for ^{255}Rf and ^{257}Db results in a value of $T_{sf} \approx$ 4000 s or $b_{sf} \approx 0.0005$ for ^{256}Db. Therefore these sf events are assigned to ^{256}Rf, produced by $^{256}\mathrm{Db} \stackrel{EC}{\rightarrow} {}^{256}\mathrm{Rf} \stackrel{sf}{\rightarrow}$. Thus we obtain a value $b_{EC} = 0.35 \pm 0.12$ for ^{256}Db.

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