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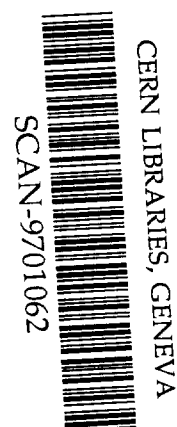
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Abstract

For the first time, chemical separations of element 106 (Seaborgium, Sg) were performed in aqueous solutions. The isotopes ^{265}Sg and ^{266}Sg were produced in the $^{248}\text{Cm} + ^{22}\text{Ne}$ reaction at a beam energy of 121 MeV. The reaction products were continuously transported by a He(KCl)-jet to the computer-controlled liquid chromatography system ARCA. In 0.1 M HNO_3 / 5×10^{-4} M HF, Sg was found to be eluted within 10 s from 1.6 x 8 mm cation-exchange columns (Aminex A6, $17.5 \pm 2 \mu\text{m}$) together with the hexavalent Mo- and W-ions, while hexavalent U-ions and tetravalent Zr-, Hf-, and element 104 ions were strongly retained on the column. Element 106 was detected by measuring correlated α -decays of the daughter isotopes 78-s $^{261}\text{104}$ and 26-s $^{257}\text{102}$. For the isotope ^{266}Sg , we have evidence for a spontaneous fission branch. It yields a partial spontaneous-fission half-life which is in agreement with recent theoretical predictions. The chemical results show that the most stable oxidation state of Sg in aqueous solution is +6, and that like its homologs Mo and W, Sg forms neutral or anionic oxo- or oxohalide-compounds under the present condition. In these first experiments, Sg exhibits properties very characteristic of group 6 elements, and does not show U-like properties.

1. Introduction

1.1. The chemical aspect

All experimental findings about the chemistry of element 104, Rutherfordium, Rf, and element 105, Hahnium, Ha, show that they constitute the first two transactinide elements*. A complete review of their chemical properties with references is given in [1]. The currently known nine transactinide elements have been placed in the Periodic Table under their lighter homologs of the 5d series Hf, Ta, W, Re, Os, Ir, Pt, Au, and Hg, see Fig. 1.

Our knowledge about the chemistry of elements 104 and 105 justifies placing Rf in group 4 below Zr and Hf, and Ha in group 5 below Nb and Ta. However, the chemistry of both elements 104 and 105 provided many surprises [1]. The non-Ta-like behaviour of Ha in aqueous solution [2–5], for example, and its similarity to Nb and/or Pa, depending on its chemical environment, demonstrated that the chemical properties cannot be reliably extrapolated from the trends observed in its lighter homologs. Such surprises have not only been observed in the aqueous-phase chemistry of element 105 but also in thermo-chromatographic [6,7] and gas-chromatographic experiments [8–11]. Some of the observed deviations from the periodicity of chemical properties based on extrapolation from lighter homologs in the Periodic Table are presumably results of relativistic effects. These have been predicted for some time [12]. The most recent atomic and molecular calculations [13–19] take the relativistic effects into account and calculate important, basic properties of atoms and molecules. Still, empirical correlations are needed to predict chemical properties from the results of these calculations. Therefore, it is of great interest to investigate to what extent the chemical properties of element 106 resemble properties of the lighter homologs in group 6, Mo and W, or rather those of the pseudo-group-6 element U.

1.2. The nuclear aspect

Recently, the isotopes ^{265}Sg and ^{266}Sg were discovered as 5n- and 4n-products from the fusion of ^{22}Ne with ^{248}Cm [20]. After separation in a gas-filled recoil-separator, ^{265}Sg was identified by correlated $\alpha - \alpha - (\alpha)$ -decay chains to the known daughter nuclei ^{261}Rf ($T_{1/2} = 78$ s, $E_{\alpha} \sim 8.29$ MeV) and ^{257}No ($T_{1/2} = 26$ s, $E_{\alpha} \sim 8.22, 8.27, 8.32$ MeV). Alpha-particles with energies 8.71 to 8.91 MeV were assigned to ^{265}Sg . A production cross-section of 260 pb was measured for

*Until unanimously accepted final names for the heaviest elements are recommended by the International Union of Pure and Applied Chemistry, the names endorsed by the Nomenclature Committee of the American Chemical Society for use in the U.S. are employed in this article without any prejudice about the priority in the discovery. It should be noted that the names kurchatovium (Ku) and nielsbohrium (Ns) were used in some of the original publications of the Dubna group for elements 104 and 105. Since 1992 nielsbohrium (Ns) is used for element 107 leaving hahnium (Ha) for element 105.

121-MeV ^{22}Ne ions. The cross-section was reported with an estimated accuracy of a factor of ~ 3 . For six event pairs, observed with a cross section of 80 pb at 116 MeV and 60 pb at 121 MeV, α -decay ($E_\alpha = 8.63 \pm 0.05$ MeV) was followed by spontaneous fission (SF). These correlations were attributed to the decay chain $^{266}\text{Sg} \xrightarrow{\alpha} ^{262}\text{Rf} \xrightarrow{\text{SF}}$. Half-lives of ^{265}Sg and ^{266}Sg could not be measured due to the lack of an initial implantation signal in the silicon detectors [20].

For ^{266}Sg , a partial α half-life of 10 to 30 s was estimated from the measured α -energies. Similarly, an α half-life of 2 to 30 s was estimated for ^{265}Sg assuming a hindrance factor between 1 and 3 [20]. A contamination with the transfer product ^{256}Fm prevented observation of possible SF branches. An upper limit of 85 % SF-branching was estimated for ^{266}Sg .

The ground-state decay properties of ^{265}Sg and ^{266}Sg [20] reveal a significantly increased stability resulting from the deformed shell at $N=162$. The experimental findings are in agreement with theoretical predictions [21, 22]. To test the theoretical predictions for the SF half-lives, it is of importance to determine the partial SF half-life for ^{266}Sg . This should be measurable on pure samples of Sg after chemical separation. Half-life measurements of ^{265}Sg and ^{266}Sg are urgently needed, complemented by more data on the α -decay energies.

The estimated half-lives of ^{265}Sg and ^{266}Sg were considered long enough to encourage a joint experimental program to chemically separate and identify the heaviest Sg isotopes with a variety of automated techniques on an atom-at-time basis. All experiments aimed at the detection of correlated decay chains from ^{265}Sg and ^{266}Sg to obtain an unequivocal signal from element 106. Gas-chromatographic separations were performed with the On-Line Gas Chemistry Apparatus, OLGA III, [23, 24], and the centrifuge system SISAK III [25, 26] was used for continuous extractions from an aqueous solution into an organic phase. Results from these experiments will be reported elsewhere. In this contribution, we discuss the first results from Sg chemistry in aqueous solution, performed as an ion-exchange chromatographic separation with the Automated Rapid Chemistry Apparatus, ARCA [27].

2. Preparatory tracer experiments

2.1. Choice of the chemical system

The ARCA [27] is a miniaturized computer-controlled liquid-chromatography system. It allows group separations of elements within only a few seconds [28] and can separate individual elements within less than 30 s [3] (see below, for a more detailed discussion of the ARCA operation in the Sg experiment). The following prerequisites are needed for a successful first experiment with Sg isotopes with ARCA: (i) The chemical system must give fast access to the Sg fraction, preferentially within less than 10 s. (ii) The eluted volume and the composition of

the aqueous solution must allow a fast sample preparation for α -spectroscopy by evaporation. (iii) A Bi and Po separation is necessary to avoid interfering α -events with energies above 8.0 MeV. These can originate with high rates from isotopes produced with very large cross sections from reactions with minor target impurities in the Pb region. (iv) A good separation of trivalent actinides is required to identify, and correctly assign, SF-events from ^{266}Sg and its daughter nucleus ^{262}Rf . (v) Elements 104 and 102 need to be separated very efficiently. This last item is especially important because, if fulfilled, then, all Rf and No isotopes, observed in the Sg fraction after chemical separation, can be presumed to be daughter nuclei of Sg precursor nuclei.

We have concentrated our preparations on the following system which is apt to meet the requirements listed above: Formation of anionic, or possibly neutral, oxo- and oxofluoride compounds in dilute nitric/ hydrofluoric acid and separation on cation exchange columns.

The formation of neutral and anionic complexes with F^- ions is a characteristic property of group 4, 5, and 6 elements, see e.g. [29,30], with distinct differences between the three groups of elements. For group 6 elements, there are indications that the formation of oxo-complexes, neutral or anionic, may prevail over the oxofluoride complexes at low F^- concentrations, see [31] and references therein. The complexing behaviour of these transition metals has been investigated on ion exchange resins, see e.g. [32–36]. It was applied to study anionic fluoride complexes of element 104 on-line with an anion exchange resin [37]. With a similar technique, preparatory experiments have been performed to separate Ta and W tracer activities on anion-exchange columns to model element 106 separations [38,39]. More recently, the behaviour on cation exchange resins has been investigated in the preparation of Sg experiments [40,41].

Batch experiments gave low distribution coefficients, K_D , ($K_D \leq 6$) for the group 6 elements Mo and W on the cation exchange resin Dowex 50W X 8 from 0.1 M HCl/ 1×10^{-4} M HF [41]. Under the same conditions, trivalent ions and the tetravalent Zr and Hf ions are strongly adsorbed with a $K_D \geq 10^3$. Increasing the HF concentration to 10^{-3} M yielded K_D -values of about four for Mo and W, and about 80 for Zr and Hf.

It was assumed that the group 4 elements form cationic complexes of the type MF^{3+} , MF_2^{2+} or MF_3^+ which are retained on the cation exchange resin. Presumably, Mo and W form anionic complexes of the type MO_3F^- , MO_2F_3^- , $\text{MO}_2\text{F}_4^{2-}$ and MOF_5^- at a sufficiently high HF concentration, and, more likely, the oxo-compound MO_4^{2-} in very dilute HF. Moreover, the formation of neutral compounds can not be excluded. Theoretical calculations on the electronic structure and redox potentials predict that, within the series of MO_4^{2-} ions formed in aqueous solution, SgO_4^{2-} will be the most stable [18]. From this we expect that in very dilute HF solutions Sg forms the SgO_4^{2-} complex which should follow the lighter homologs Mo and W in the chemical separation. While K_D -values of about 100, observed for Mo and W on the anion exchange resin Dowex 1X8

from a solution of 0.1 M HCl with 10^{-3} M and 10^{-4} M HF [41], support the assumption of anionic complexes, we cannot totally exclude the formation of the neutral species MO_2F_2 . In the tracer experiments, a predominant formation of fluoride complexes, instead of chloride complexes, can be assumed because of the much higher complex stability of the fluorides. To avoid any competition from the chloride anions in the complex formation, we decided to substitute 0.1 M HNO_3 for 0.1 M HCl in the Sg experiments. We have experimentally verified that the results obtained in batch experiments with 0.1 M HCl [41] are also obtained in 0.1 M HNO_3 [42].

2.1. On-line tracer separations

Carrier-free tracer activities were obtained as fission products (^{93}Y , ^{97}Zr , ^{99}Mo) from the Mainz TRIGA reactor, and from the reaction of 155-MeV ^{20}Ne ions with enriched ^{152}Gd and ^{nat}Sm targets. The latter experiments, performed at the PSI Philips-cyclotron and at the GSI UNILAC, produced tracers activities of ^{162}Lu , ^{165}Hf , and ^{168}W . A He(KCl)-jet system, similar to the one described below for the Sg experiment, was applied in these experiments to transport the tracers to the ARCA [27]. After typical collection times of 120 s, tracer activities were dissolved from the collection position on the top slider of ARCA and were washed onto the 8×1.6 mm chromatographic column filled with the cation exchange resin Aminex A6, 17.5 ± 2 μm . Elutions were performed with 1 ml/min flow rate, and fractions were collected to monitor chemical yields, purities of the chemical separations, and elution curves by applying X-ray and γ -spectroscopy.

Earlier experiments have indicated that some of the transition metal ions are strongly adsorbed on surfaces of commonly used materials. The first aqueous chemistry experiment on element 105 is one of the examples [2]. Therefore, we have investigated the sorption behaviour of Hf and W activities on various possible materials for the top slider in ARCA. W was most efficiently dissolved from Ti sliders with the solutions envisaged for the Sg experiments. For example, in 0.1 M $\text{HNO}_3/5 \times 10^{-4}$ M HF 66 % of the W was dissolved and washed through the ARCA (columns were empty) within 5 s. This includes about 2.5 s dead time from the dead volume in ARCA. It should be noted that W and Hf were strongly adsorbed on some of the tested materials, like stainless steel, polymethacrylate (PMMA, Plexiglas), polyetheretherketone (PEEK), and polychlorotrifluorethylene (PCTFE, Kel-F). The result was also poor for all materials when pure 0.1 M HCl with no additional HF was applied.

In our elution experiments, we kept the HNO_3 concentration constant at 0.1 M and varied the HF concentration between 10^{-4} M and 10^{-3} M. Fig. 2 shows the results for W and Hf elutions with 0.1 M $\text{HNO}_3/1 \times 10^{-3}$ M HF (upper panel), and with 0.1 M $\text{HNO}_3/5 \times 10^{-4}$ M HF (lower panel). Either eluent dissolves and elutes more than 85 % of the W within 10 s. The fluoride concentration in 0.1 M $\text{HNO}_3/1 \times 10^{-3}$ M HF is obviously high enough to form neutral or anionic

complexes of Hf which appear in the W fraction (upper panel of Fig. 2). Reducing the HF concentration to 5×10^{-4} M provides conditions for a clean separation of group 6 elements (lower panel in Fig. 2). No di- or trivalent actinides and no group 4 elements are eluted within the first 15 s.

3. Seaborgium experiments

3.1. Production, transport, and separation of $^{265,266}\text{Sg}$

To produce ^{265}Sg and ^{266}Sg , we used the ^{22}Ne on ^{248}Cm reaction, as in the discovery experiment [20]. All targets were prepared by electrodeposition techniques. In a first and shorter series of experiments (1172 separations with a total dose of 2.31×10^{17} ^{22}Ne ions) we used targets of about $150 \mu\text{g}/\text{cm}^2$ ^{248}Cm . A target with a thickness of $950 \mu\text{g}/\text{cm}^2$ ^{248}Cm was used in the second, more sensitive experiment (3900 separations with a total dose of 5.48×10^{17} ^{22}Ne ions). The isotopic composition of the latter target was 95.7 % ^{248}Cm and 4.2 % ^{246}Cm . During all experiments, the 153.8-MeV beam from the UNILAC was degraded in energy by a $2.67 \text{ mg}/\text{cm}^2$ Be vacuum-window, $0.65 \text{ mg}/\text{cm}^2$ N_2 cooling-gas, and the $2.65 \text{ mg}/\text{cm}^2$ Be target-backing before the ^{22}Ne entered the target at 122 MeV. The energy in the middle of the target was 121 MeV. A good agreement was achieved between the calculated energy loss and a measurement of the energy loss using a Passivated Ion-implanted Planar Silicon (PIPS) detector. For this measurement, the calibration was done with a 122-MeV ^{22}Ne -ion beam from the UNILAC. The beam intensity was low enough not to damage the detector. Again, 122 MeV was measured after completing the experimental set-up, and increasing the ^{22}Ne -ion beam energy from the UNILAC to 153.8 MeV. The beam current in the Sg experiment was about 0.5 particle microamperes ($3 \times 10^{12} \text{ s}^{-1}$).

A bombarding energy of 121 MeV (middle of the target) was chosen, because HIVAP calculations [43] show, that the excitation function for the production of ^{265}Sg peaks in this energy region. The data from the discovery experiment [20] support this assumption. In addition, it was hoped that the 4n-channel leading to ^{266}Sg , may still have a cross section high enough to observe α -SF correlations from the decay of ^{266}Sg .

The reaction products recoiling out of the target were stopped in He gas (about 1.7 bar) which had been loaded with KCl aerosols produced by sublimation from the surface of KCl powder at 640 °C. The activity, attached to the aerosols, was swept out of the recoil chamber with the He gas (2.5 l/min) into a capillary (2 mm i.d.). To avoid losses of charged aerosols, stainless steel capillaries were used to transport the products about 18 m to the collection site in ARCA. There, the He gas with the aerosols exited into the collection site of ARCA through a nozzle. The He was pumped away perpendicular to this flow direction. The KCl-aerosols, together with the reaction products, were collected in forward direction

by inertial impactation on one of the two dips (0.5 mm deep, 5 mm in diameter) in the top slider of ARCA [27]. This new technique provided a better and more reliable deposition yield and jet operation compared with the collection on the polyethylene frits used in earlier experiments. In the main Sg experiments, the activity-bearing aerosols were collected for 45 s (60 s in the experiments with the 0.15 mg/cm² target) and, after end of the collection time, the collection site was moved into position for chemical processing of the activity.

The He(KCl)-jet transport was optimized in a number of preparatory experiments [44–46]. To determine the transport yield of products from heavy-ion reactions, we first collected the recoils directly behind the targets in a catcher foil to get a reference value (100 %). Products transported in the jet were collected on a glass fiber filter, and, as for the catcher foil, X-ray and γ -spectroscopy were performed. By comparing the apparent production rates measured after transport through the He(KCl)-jet with the absolute production rates from the catcher foil, the transport efficiency was determined. We also measured the aerosol size distribution and concentration for each set of parameters to determine correlations with the transport yield. These experiments showed that, under optimized conditions, a stable transport yield of about 50 % can be obtained over periods of many days. The He(KCl)-jet transport efficiency was again checked with ¹⁶⁹W, produced in ²²Ne on ¹⁵²Gd, before the Sg experiment began, and a yield of 45 % was obtained. The transport efficiency was measured frequently with two different set-ups during the experiment. This was done by collecting the KCl-aerosols from the jet for 60 s either on a nucleopore filter or in a collection site similar to the top part of ARCA. The production rate of ^{252–255}Fm transfer products was determined by α pulse-height analysis, and was compared with the known cross section of about 3 μ b for ²⁵⁴Fm and 1 μ b for ²⁵⁵Fm [47]. Collecting the aerosols on a nucleopore filter gave transport yields of 50 % and higher, while a transport and collection yield of 45 % was obtained with the set-up which simulates the condition in ARCA. All yields stayed constant over the time of the experiment. A yield of 45 % was assumed in the data analysis of the Sg experiment. From the void volumes in the jet and the He flow rate we calculated a transport time of 3 s into ARCA.

For the chemical separation of Sg in ARCA, we chose the 0.1 M HNO₃/5 \times 10⁻⁴ M HF solution, as described in Section 2. The HF concentration in this solution is low enough to achieve a good separation from group 4 elements, and we could perform two separations on each column in ARCA without a contamination from unwanted species. In our search for Sg, we collected the eluent from the first 10 s on a Ta disk. As the mean time for the chemical separation of Sg from Rf and No we took 5 s (after end of collection), see Fig. 2. The evaporation of the eluent started with the beginning of the elution [27]. For a faster and smoother evaporation, as compared with our previous techniques, where little droplets were collected [27], we continuously sprayed the eluent through a 60 \pm 2 μ m i.d. nozzle onto the Ta disks. An additional \approx 20 s were needed after the end of elution to

prepare a sample for α -particle spectroscopy and SF fragment-energy measurements. On the average, measurements started 38 s after end of collection. The overall chemical yield, which includes dissolving the products, chemical separation on the cation exchange column, and sample preparation, was determined to be 80 % in a large number of preparatory experiments with Mo and W tracers.

3.2. Detection of α and SF activities

α -particle and SF fragment pulse-height analyses were performed on each sample for 360 s (480 s for the experiments with 60 s collection time) using a system of eight 450 mm² PIPS detectors. The energy of each event with the time after start of counting and detector identification was stored in list mode on magnetic disk and tape. The energy resolution, which was determined by the sample quality, was about 60 keV (FWHM) for α -particle energies between 5 and 12 MeV. The energy calibration over all detectors was accurate to about 10 to 20 keV for α -particles. An approximate fission-energy calibration was obtained by extrapolating the α -energy calibration. The detector efficiency for the detection of α -particles was 33 %, implying 66 % efficiency for the detection of a fragment from SF decay. From this we calculate an efficiency of 11 % for the detection of an α - α correlation and 22 % for an α -SF correlation from parent-daughter decays.

4. Nuclear properties of ^{265,266}Sg and daughter nuclides

Let us first recall what is known about the decay of ²⁶⁵Sg and ²⁶⁶Sg. The discovery experiment assigned α -particles with energies of 8.71, 8.81, 8.85, and 8.91 MeV to the decay of ²⁶⁵Sg and energies of 8.54, 8.59, 8.60 (2 events), 8.69, and 8.74 MeV to the decay of ²⁶⁶Sg [20]. Most detector strips of this experiment had an energy resolution of about 100 keV [20]. The six observed α -decays of ²⁶⁶Sg were followed by SF with correlation times which gave a half life of (1.2 +10/-0.5) s for ²⁶²Rf. Recently, a (2.1 ± 0.2)-s SF activity has been identified in the ²²Ne + ²⁴⁴Pu reaction and assigned to ²⁶²Rf [48]. For the moment, it remains an open question whether the SF activity with $T_{1/2} \approx 47$ ms, which was tentatively assigned [49] to ²⁶²Rf, belongs to an isomeric state in ²⁶²Rf or to a different nuclide. In fact, an unassigned 1.3-s SF activity was also produced with a similar cross section in the experiment which yielded the 47-ms component [49]. The α -decay of ²⁶⁵Sg is followed by the α -decays of 78-s ²⁶¹Rf ($E_\alpha=8.28$ MeV) and 25-s ²⁵⁹No ($E_\alpha=8.22$, 8.27, and 8.32 MeV).

Two α -decay chains into ²⁶⁵Sg were observed in the most recent discovery of element 112 [50]. Life times of 7.4 and 24.1 s were measured for the two observed decays of ²⁶⁵Sg. An α -energy of 8.77 MeV was determined in one of the ²⁶⁵Sg

decays while the second α -particle left the detector and only a partial energy of 4.6 MeV was registered. It is interesting to note that in one of the decay chains, an α -particle with a hitherto unobserved energy of 8.52 MeV was assigned to ^{261}Rf . This α -energy is in good agreement with Q-value systematics [51]. It may have escaped observation in earlier experiments because this energy coincides with one of the α -particle energies of ^{212m}Po , a contamination present in most experiments.

Decay data, measured in a companion experiment to ARCA with an on-line gas-chromatographic technique [24], yielded in a preliminary analysis three α - α correlations and one α -SF correlation [24]. The mother events were assigned to decays of ^{265}Sg ($E_\alpha=8.84, 8.85, \text{ and } 8.86$ MeV with life-times of 27.3, 0.6, and 2.8 s, respectively) and to ^{266}Sg ($E_\alpha=8.56$ MeV with a life-time of 48.9 s). For ^{265}Sg , a half-life of $(7.1 +8.6/-2.5)$ s was obtained with a 68 % confidence interval (c.i.) $(+ 27/-4.1$ for a 95 % c.i.). If we include in the analysis the life-times of ^{265}Sg observed in the two decay chains of $^{277}\text{112}$ [50], a half-life of $(8.6 +6.5/-2.6)$ s results $(+ 18/-4.4$ for a 95 % c.i.). From the one decay of ^{266}Sg , a half-life of $(34 +163/-15)$ s was obtained with a 68 % c.i. $(+1306/-27.4$ for a 95 % c.i.).

5. Results and discussion

5.1. ARCA – ^{265}Sg

In the ARCA experiment, 3900 collection and elution cycles with the time sequence given in Section 3 were run to get the α -energy spectrum shown in Fig. 3. A small contamination of $^{211m,212,213}\text{Po}$ and ^{214}At prevents assigning α 's from this spectrum to the decay of Sg or its daughter nuclei without further information. A time-distribution analysis of the α -events between 8.0 and 9.0 MeV does not show any significant decay over the measuring time of 6 min indicating relatively long-lived precursors of ^{213}Po , ^{212}Po , and ^{214}At . Possible candidates are $^{212}\text{Bi/Pb}$, $^{213}\text{Bi/Pb}$, and ^{226}Pa . The latter nuclide has been clearly identified by triple $\alpha - \alpha - \alpha$ -correlations; e.g. an α -particle with an energy of 6.76 MeV (^{226}Pa) observed 65 s after the start-of-measurement, followed 7 s later by an α -particle with 6.94 MeV (^{222}Ac) and 3 ms later by one with with 8.86 MeV (^{214}At). The observation of triple correlations in the background was used as one check to assure the proper functioning of the data acquisition.

To identify α -decays of ^{265}Sg and the daughter nuclei ^{261}Rf and ^{257}No , we have searched for correlated α - α decays. All correlated α - α events with energies between 6 and 10 MeV are depicted in Fig. 4. Two energy windows were applied in our search.

We have searched for the $^{265}\text{Sg} \xrightarrow{\alpha} ^{261}\text{Rf} \xrightarrow{\alpha}$ decay in the so-called "parent-daughter" window ("P" in Fig. 4). For the first ("parent") α -particle, possibly arising from a ^{265}Sg decay, we accepted energies between 8.50 and 9.10 MeV

(on the abscissa or E1-scale in Fig. 4). For the second ("daughter") α -particle the energy window was open between 8.10 to 8.60 MeV (on the ordinate or E2-scale in Fig. 4) to include α -particles from the decay of ^{261}Rf and ^{257}No . One correlated event with E1 = 8.52 MeV (Δt_1 (time after-start-of-measurement) = 142 s) and E2 = 8.12 MeV (Δt_2 (time after first α) = 3.0 s) was observed in the "parent-daughter" window.

To judge the significance of this one event for being a true decay of ^{265}Sg followed by the decay of ^{261}Rf , we have calculated the number of expected random events by a Monte-Carlo simulation of our experimental data. 1565 measured single α -events between 6 MeV and 12 MeV were used as a data base for randomly drawing pairs of energies together with the respective times, T_{meas} , when these events had appeared. The times were converted into random times, T_{simu} , by $T_{rand} = -T_{meas} \times \ln(RND)$ with random numbers, RND , between 0 and 1. This yields, for each individual event, an exponential time-distribution which reflects the most probable life-time of this event. 4×10^7 α - α -correlations were simulated, a factor of 10^5 more than the measured 407 correlated α - α -decay events.

The Monte-Carlo simulation gives a random rate of 0.56 correlations (expectation value, μ , in Table 2) for the "parent-daughter" energy window, where we have measured one correlated α - α decay. For an expectation value of 0.56 the probability is 32.1 % for observing one random correlation, see Table 2. From this we conclude, that we have no evidence for the direct observation of a ^{265}Sg α -decay.

In a second energy region, we have looked for the $^{261}\text{Rf} \xrightarrow{\alpha} ^{257}\text{No} \xrightarrow{\alpha}$ decay ("daughter-daughter"). For the first α -particle, presumably arising from a ^{261}Rf decay, we have investigated two options. Option A is based on the assumption that ^{261}Rf has only one characteristic α -decay with $E_\alpha=8.28$ MeV. In this option (A) energies between 8.20 and 8.36 MeV are accepted for the first α . Option B is based on the assumption that ^{261}Rf has a second characteristic α -decay energy with $E_\alpha=8.52$ MeV [50]. In this option (B) α -particles with energies between 8.20 and 8.60 MeV are viewed as possible candidates for the decay of ^{261}Rf . For the second α -particle, the decay of ^{257}No , an energy window from 8.10 to 8.40 MeV was selected. To determine these limits, we added (subtracted) 80 keV to the highest (lowest) known α -decay energy of the respective nucleus to take into account the energy resolution in our experiment and the uncertainty of the energy calibration.

The two correlated α - α decays observed in the energy window "DA" (option A) of Fig. 4 are listed in Table 1 as events # 1 and 2. One correlation started with a 8.24 MeV α -particle 33.5 s after start-of-measurement followed by a 8.22 MeV α -particle 67.4 s after the first α . The second correlation was a 8.26 MeV α -particle 22.7 s after the start-of-measurement followed after 8.2 s by an event with 8.10 MeV. A third correlated α - α decay (event # 3) was observed in the larger energy window "DB" of Fig. 4 when option B is applied. This event is identical with the one we already discussed in the "parent-daughter" energy

window.

A comparison of these numbers of measured events with the expected random correlations, as obtained from the Monte-Carlo simulation, shows that the observed $\alpha - \alpha$ -correlations are very likely true α -decays of the ^{265}Sg -daughter nuclei ^{261}Rf and ^{257}No . The two events from option A have to be compared with an expectation value of 0.15 for random correlations. This leads to a probability of only 0.96 % that the two events are random, see Table 2. Applying option B we have to compare three measured events with an expectation value of 0.27 for random correlations. This gives an even lower probability of only 0.24 % that the three events are random correlations, and a high confidence that we have measured the decay of ^{261}Rf and ^{257}No as the daughter nuclei of ^{265}Sg . Isotopes of Rf and No can only be observed as the decay products of Sg because they were chemically separated on the cation exchange column in ARCA. Most likely, the decay of ^{265}Sg was not seen because it decayed in the time interval between the end-of-separation and the start-of-measurement. It could have also escaped the detector.

From the ARCA experiment, we are not able to determine a half-life for the decay of ^{265}Sg . A calculation of cross sections, based on the observed decay of the ^{265}Sg daughter nuclei, versus an assumed half-life is shown in Fig. 5. The 68 % c.i. band shown in Fig. 5 has been determined with a maximum likelihood method, which was applied in our data analysis to determine confidence intervals for expectation values between zero and nine. Included in Fig. 5 is also the result from the OLGA experiment [24]. Combining the results from both experiments considerably reduces the range of possible $T_{1/2}$ - and σ -values, see Fig. 5. These results are in good agreement with half-lives deduced from the observed α -energies in the discovery experiment [20] and with the cross sections, which were reported with an uncertainty of a factor ≈ 3 . The measured cross sections are somewhat higher than the ones calculated with HIVAP [43], but with the present error bars this does not constitute a significant difference.

5.2. ARCA – ^{266}Sg

To identify decays of ^{266}Sg , we have searched for correlated α -SF events from the α -decay of ^{266}Sg followed by a SF from ^{262}Rf , and we have analysed single SF-fragment events which may originate from a SF-decay branch in ^{266}Sg . No α -SF correlation ($7 \text{ MeV} \leq E_\alpha \leq 10 \text{ MeV}$) was observed in the ARCA experiment, and we have measured nine single SF-events, see Table 3, after 3900 experiments performed with the 0.95 mg/cm^2 ^{248}Cm -target. An analysis of the time distribution of these nine SF-events yields a half-life of $(94 +46/-23) \text{ s}$ (68 % c.i. following the formalism given in [52]) if a decay of one component is assumed. In the following, we will discuss possible sources of SF-fragment background which may have contributed to the observed effect. This can be estimated from known isotope ratios, e.g., the isotopic composition of the target and the production cross sec-

tion of Fm-isotopes. In this procedure, we have used the characteristic α -decay of a leading isotope to determine its amount and to deduce the decay rate of the isotope with SF-decay.

The group of α -particles with an energy of about 7.2 MeV, see Fig. 3, decays with a half-life which is in very good agreement with 25.2 s, the known half-life of $^{211\text{m}}\text{Po}$. A decay analysis for a potentially longer-lived component in the energy region 7.14 to 7.29 MeV results in a maximum of two observed α -particles for the sum of all experiments. A small ^{254}Fm ($E_\alpha=7.19$ and 7.15 MeV) contamination in the Sg fraction could have been the origin. From known decay properties and the cross-section ratios of the transfer products $^{254}\text{Fm}/^{256}\text{Fm}$ [47] we calculate a total of $(0.24 +0.76/-0.24)$ SF-fragments (95 % c.i.) from the decay of ^{256}Fm in the 3900 ARCA experiments.

A second potential source for SF-fragments are small amounts of ^{248}Cm -target material knocked over into the gas-jet by Ne-projectiles. Here, the leading isotope with the strongest line in the α -energy spectrum is ^{246}Cm ($E_\alpha=5.39$ MeV). In the Sg-experiment, we have measured 31 α -particles in the energy region between 5.31 MeV and 5.47 MeV for a counting time of 467 h. During 5856 h of background measurements with blank sample plates, we have observed 283 α -particles in the same energy window. From this we calculate that 8 α -particles may originate from a ^{246}Cm contamination in the α -energy spectrum of the Sg-fractions. With the known Cm-isotope ratio and the SF-branching ratio of 8.26 % for ^{248}Cm we conclude (94 % c.i.) that the number of SF-fragments from ^{248}Cm is $(0.41 +0.59/-0.41)$.

The SF background from cosmic-rays, electronic noise etc. was estimated with a 68 % c.i. to $(1.35 +0.65/-1.35)$ SF-events $(+1.65/-1.35)$ with 95 % c.i.) from a long background measurement after the experiment.

After subtracting all background components, we calculate with a 68 % c.i. that $(7.0 +4.0/-2.2)$ observed SF-fragments $(+7.9/-4.3)$ with 95 % c.i.) remain as possible candidates for the SF-decay of ^{266}Sg . This corresponds to a cross section of $(82 +46/-26)$ pb $(+93/-51)$ pb with 95 % c.i.) if a half-life of 34 s is assumed. As we have not observed the α -decay of ^{266}Sg in the ARCA experiment, we are not able to determine directly a branching ratio for the decay. However, the SF-branch of ^{266}Sg can be obtained, with a very large error bar, from the ratio between the partial SF-decay cross-section of $(82 +46/-26)$ pb, as determined in the ARCA experiment, and the partial α -decay cross-section of $(50 +115/-15)$ pb (68 % c.i.) as reported in the OLGA experiment [24]. The ratio of the cross-section probability distributions for the α -decay and the SF-decay yields a most probable SF-branch of 62 % with an upper limit of 84 % and a lower limit of 38 % (68 % c.i.). These limits are 87 % and 22 %, respectively, when calculated for a 95 % c.i..

If we assume a total half-life of 34 s and a branching ratio of 62 %, we calculate a partial SF half-life of 55 s for ^{266}Sg . The determination of the error bar on the partial SF half-life is complicated mainly by two facts: (i) the error bars on the

total half-life and the branching ratio are strongly asymmetric, and, more critical, (ii) the SF branching-ratio in our analysis depends on the assumed half-life. For a total half-life of 34 s, the 68 % c.i. in the branching ratio yields an upper limit of 90 s and a lower limit of 40 s for the partial SF half-life. If we use an extreme approach and calculate the limits for the partial SF half-life from the limits of the 68 % c.i. of the total half-life and the 68 % c.i. limits of the branching ratios calculated for the shortest (19 s) and the longest (197 s) half-life, we obtain 518 s for the upper and 22 s for the lower limit of the partial SF half-life.

Fig. 6 shows experimentally determined partial α and SF half-lives in comparison with those from different theoretical approaches. For ^{266}Sg , the partial SF half-life of (55 +463/-33) s yields a partial α -decay half-life of (90 +1140/-59) s. The calculated half-lives of [21, 22] are in good agreement with the experimental values. The theoretical approach applied by [53] considered two possible paths into the fission valley, the so called "Old Path" and the "New Path". The "New Path", which was expected to dominate [53], can be ruled out from the experimental results. A much too short SF half-life of about 0.1 ms was predicted. For the "Old Path", however, a SF half-life was calculated [53] which is for a conservative estimate with 95 % c.i. still within the error bar of the experimental value.

5.3. Seaborgium chemistry

From the observation of the three correlated α -decay chains of Sg daughters we conclude, that, **for the first time, a chemical separation of element 106 was performed in aqueous solution.** Seaborgium shows a behaviour typical for a hexavalent element located in group 6 of the Periodic Table below Mo and W, see Fig. 1. Presumably, Sg forms the anionic oxo-compound $[\text{SgO}_4]^{2-}$ or a neutral complex. From our experiment we cannot exclude the formation of other kinds of anionic compounds, like $[\text{SgO}_3\text{F}]^-$, $[\text{SgO}_2\text{F}_3]^-$ or $[\text{SgO}_2\text{F}_4]^{2-}$. However, for the lighter homologs Mo and W, these species are only observed at higher F^- -concentrations. Contrary to element 105 in group 5 [3], where a striking similarity of its chemical behaviour to that of the pseudo-group-5 element Pa was observed, these first experiments do not indicate a Sg behaviour similar to that of the pseudo-group-6 element U, which is strongly retained on the column as UO_2^{2+} under the present conditions.

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Table 1: ^{265}Sg -events from the ARCA experiment with 121-MeV ^{22}Ne on ^{248}Cm .

Event #	E(1) (MeV)	$\Delta t1$ (s)	E(2) (MeV)	$\Delta t2$ (s)
1	8.24	33.5	8.22	67.4
2	8.26	22.7	8.10	8.2
3	8.52	142.0	8.12	3.0

E(1): energy of the first α -particle
E(2): energy of the second α -particle
 $\Delta t1$: time after start-of-measurement
 $\Delta t2$: time after 1st α -event

Table 2: Probability $P(\mu;n)$ to observe a discrete number, n , of random correlations for a given expectation value, μ , of a Poission distribution. μ is from a Monte-Carlo simulation of the experimental data for a given energy window, Win (see text and Fig. 4).

Win	μ	$P(\mu;n) / \%$			
		n=0	n=1	n=2	n=3
P	0.56	57.0	32.1	8.95	1.65
DA	0.15	86.1	12.9	0.96	0.05
DB	0.27	76.3	20.7	2.77	0.24

Table 3: Single SF-events from the ARCA experiment.

Event #	Fragment energy (MeV)	Δt (s)
1	27	60.4
2	30	161.8
3	31	97.9
4	55	131.6
5	91	0.9
6	103	180.1
7	105	118.2
8	108	367.8
9	110	102.4

Δt : time after start-of-measurement

Figure Captions

Fig. 1. Periodic Table of the Elements. The arrangement of the actinides reflects that the first actinide elements still resemble, to a decreasing extent, the chemistry of the other groups: Th the fourth group below Hf, Pa the fifth group below Ta, and U the sixth group below W. The known transactinide elements 104 through 112 shall take the positions below Hf in group 4 and Hg in group 12. Element 106, Seaborgium, is placed in group 6.

Fig. 2. Elution curves for W-tracer, modeling the Sg separation on ARCA, and of Hf-tracer, modeling the Rf behaviour, in 0.1 M HNO₃/1×10⁻³ M HF (upper panel), and in 0.1 M HNO₃/5×10⁻⁴ M HF (lower panel) solution. Elutions were performed with a flow rate of 1 ml/min from the 1.6×8 mm columns in ARCA filled with the cation exchange resin Aminex A6.

Fig. 3. α -energy spectrum of 3900 chemically separated Sg fractions observed in the bombardement of ²⁴⁸Cm (0.95 mg/cm²) with 121-MeV ²²Ne ions.

Fig. 4. Correlated α - α events observed in individual Sg fractions after chemical separation with ARCA. E1 represents the α -energy of the α -particle observed first, E2 the energy of the subsequent second α -particle. The energy windows are given for our search for the ²⁶⁵Sg $\xrightarrow{\alpha}$ ²⁶¹Rf $\xrightarrow{\alpha}$ decay ("P"), and for the ²⁶¹Rf $\xrightarrow{\alpha}$ ²⁵⁷No $\xrightarrow{\alpha}$ decay ("DA" and "DB").

Fig. 5: Cross section of ²⁶⁵Sg as a function of half-life from the ARCA experiment (solid line) with a 68 % c.i. (dashed lines). In addition, the cross section from the OLGA experiment (dot) [24] is included. The error bars (68 % c.i.) reflect the statistical error. The dashed region indicates the area of possible T_{1/2} - and σ - values that are compatible both with the OLGA and the ARCA results.

Fig. 6: Measured (solid symbols: squares for SF, dots for α -decay) and calculated partial half-lives (open symbols) for isotopes of element 106. The experimental data for ²⁶⁶Sg are from this work and from [24]; see text for the discussion of the error bars. Calculations are from [21,22] for α -decay (open circles) and SF (open squares). A different theoretical approach was used in [53] to calculate SF half-lives (open triangles) for two different pathes into the fission valley.

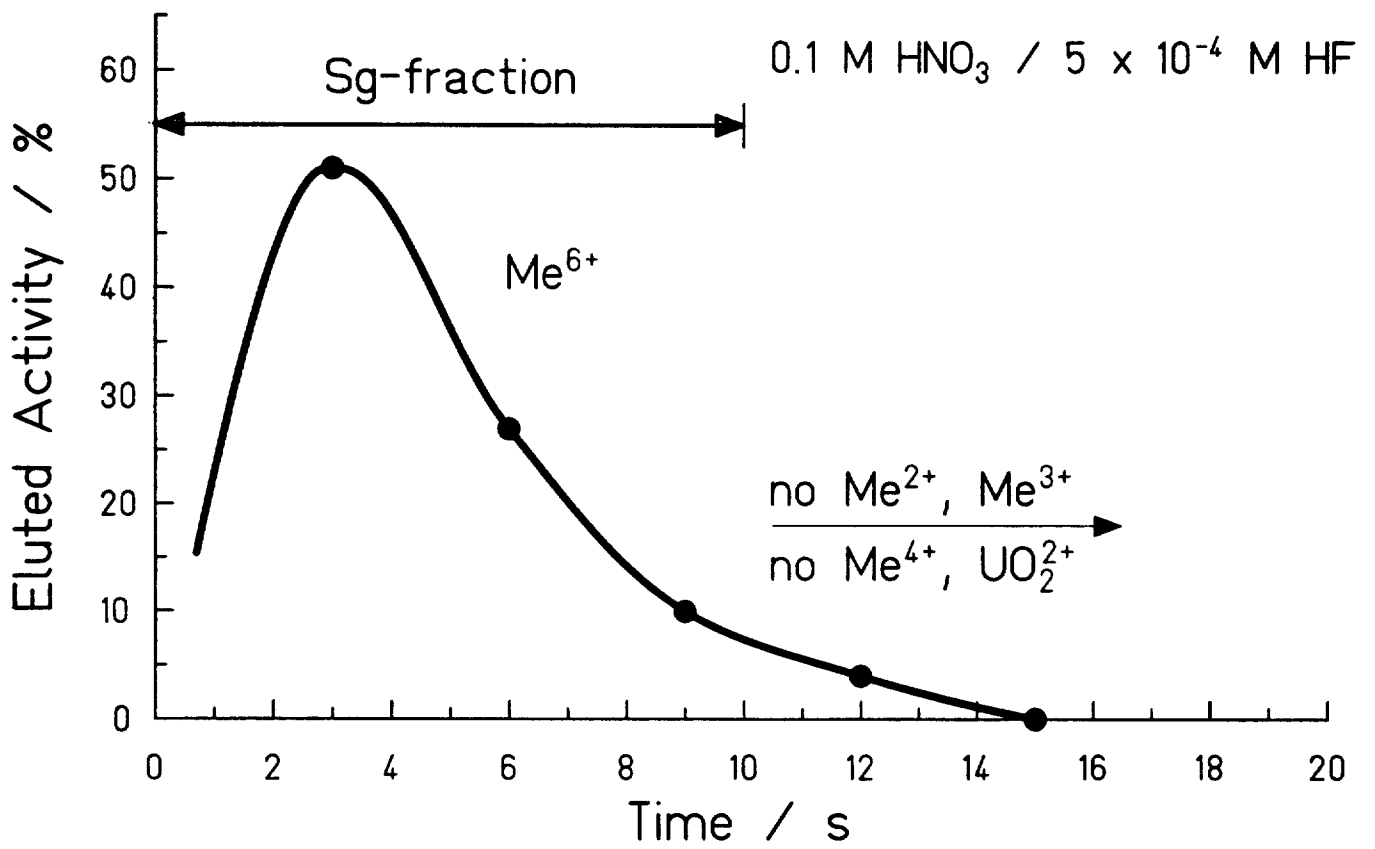
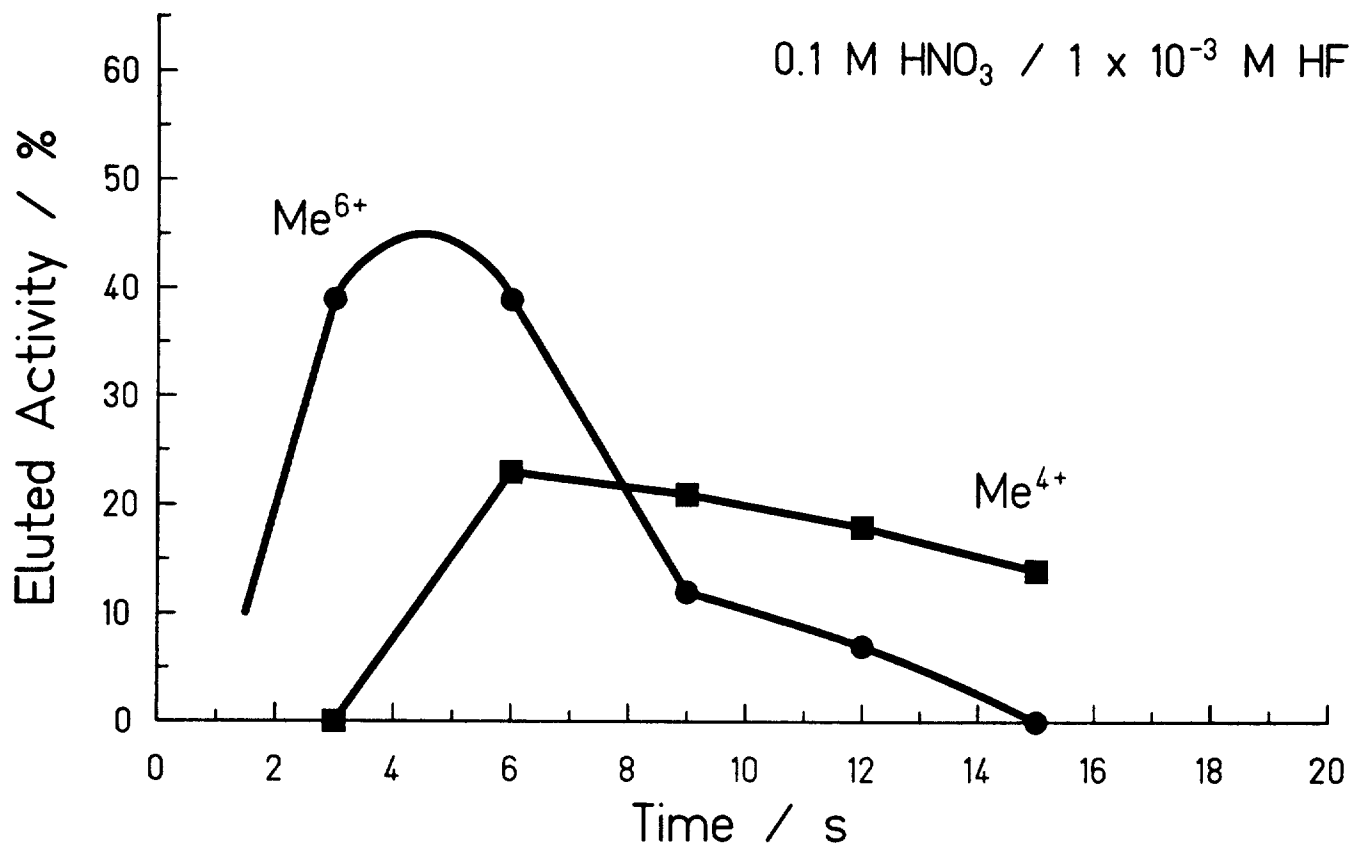


Fig. 2

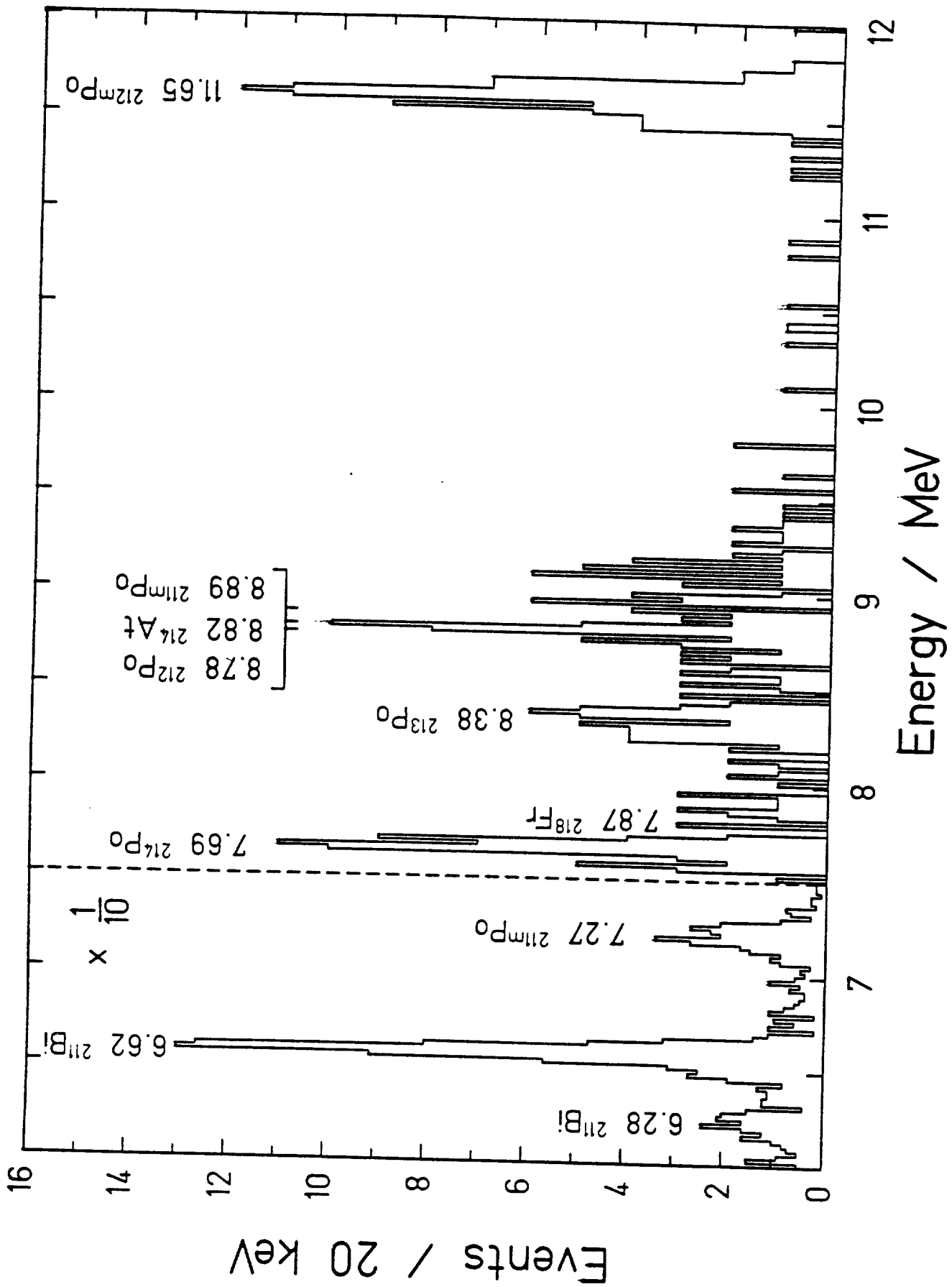


Fig. 3

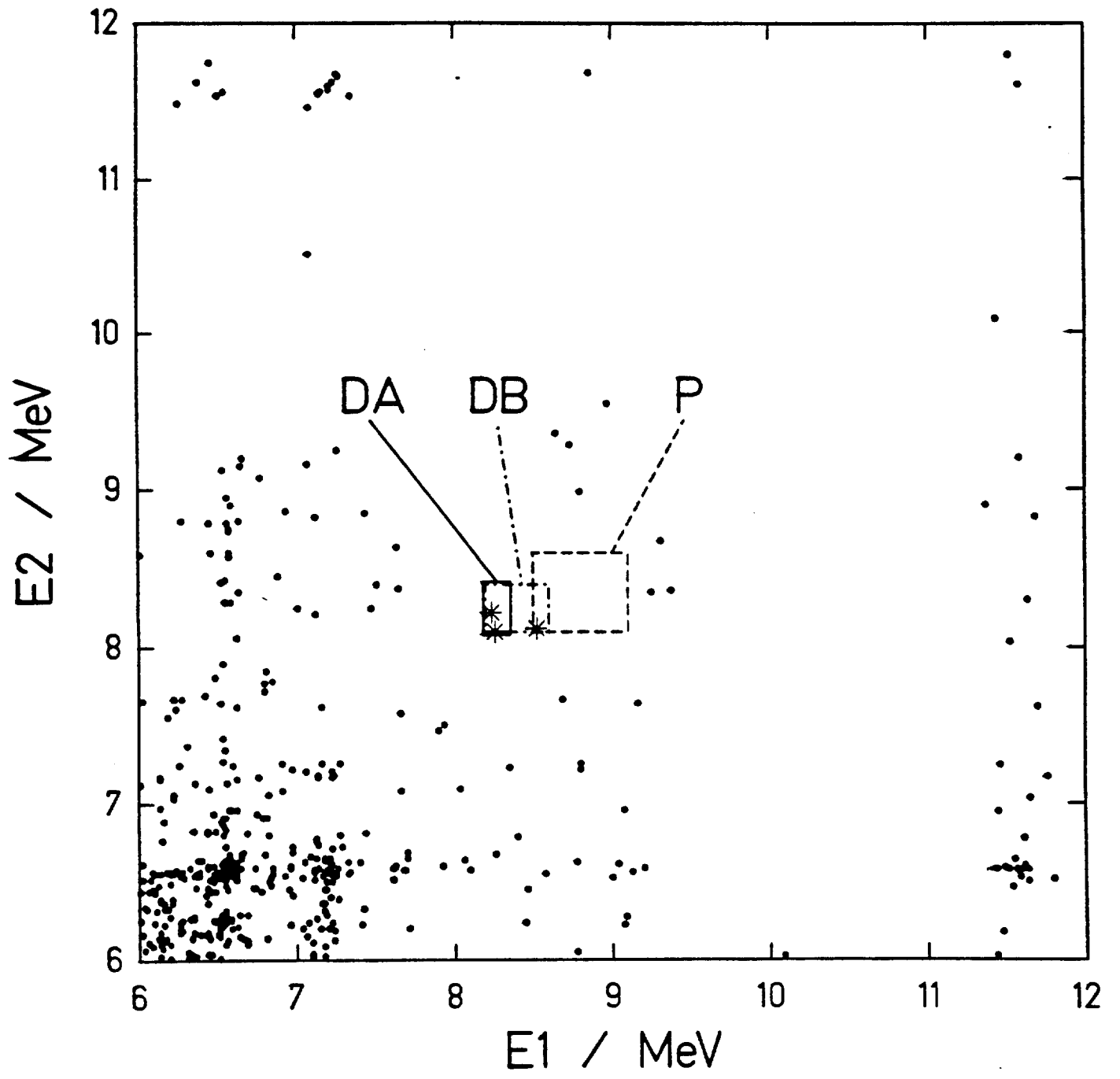


Fig. 4

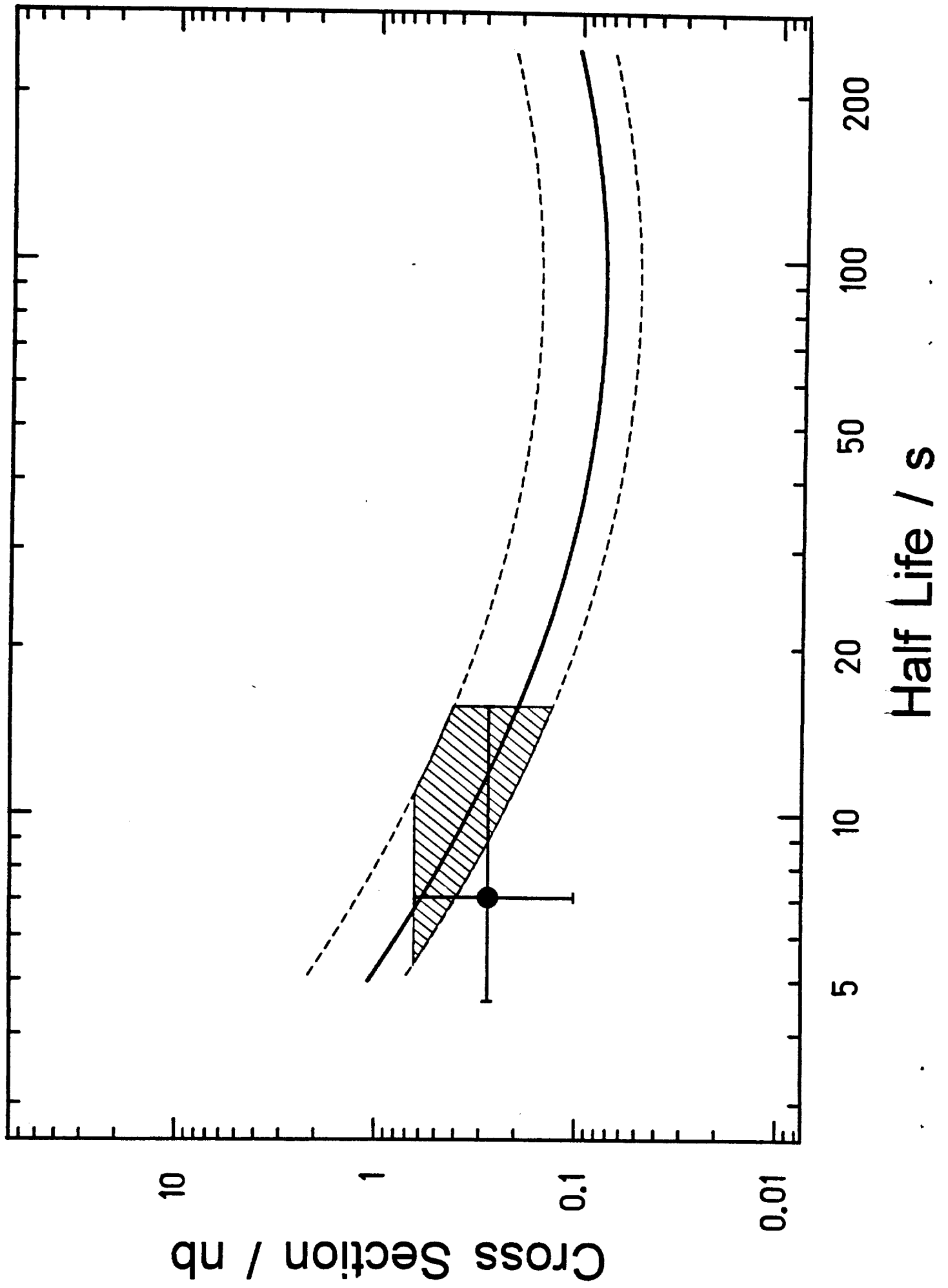


Fig. 5

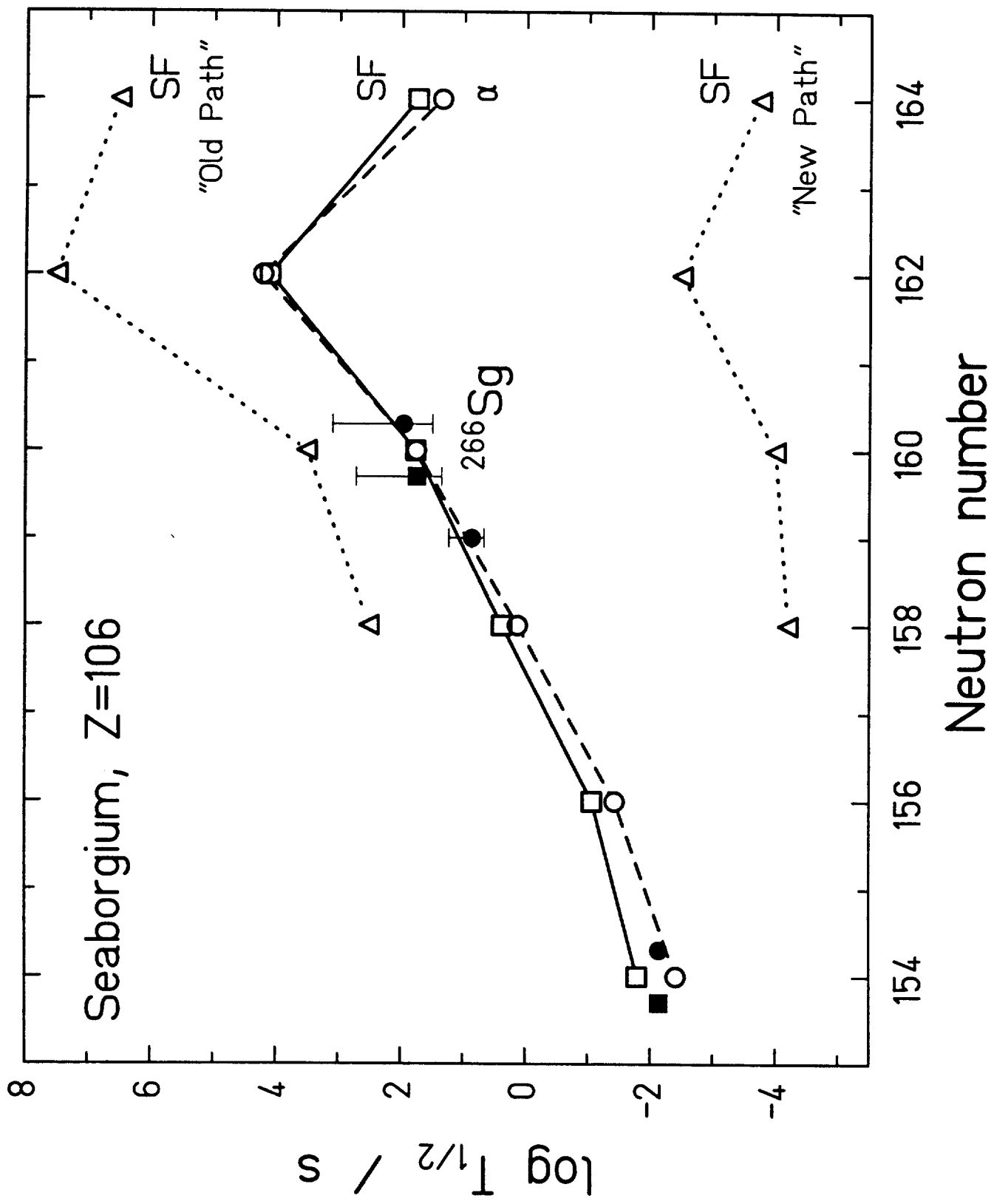


Fig. 6

