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Plans to identify heavy elements produced in reactions with cross-sections of 1 pb and below

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Abstract

Reactions for the production of heavy elements have been extensively studied since universal heavy ion accelerators became available. New separation and detection systems have been developed, leading to the identification of the new elements $_{107}$Ns, $_{108}$Hs and $_{109}$Mt. Based on the results obtained, estimates for the production of still heavier elements are given. These extrapolations demand enough experimental sensitivity to measure nuclei produced in reactions of cross-section 1 pb. Similar sensitivity is necessary to measure small decay branches of already known nuclei, to investigate fine structure and to search for new, more neutron-deficient or neutron-rich isotopes. Their stability is determined by details of the nuclear shell structure. Measured data on the reaction processes and the decay properties of the nuclei produced are the bases for a better physical understanding of the formation and stability of the heaviest nuclei. Experimental developments are described aiming at identifying nuclei and their radioactive decays on a cross-section level of 1 pb.

1. Introduction: why picobarns?

In earlier cross-section compilations of nuclear fusion reactions, such as that published by Neubert [1] in 1973, few of the collected data were in the range close to 1 nb. Since then, more powerful accelerators have come into operation: in Berkeley, the SuperHILAC; in Dubna, the U400 cyclotron; and, in Darmstadt, the UNILAC. In all three laboratories, investigations to search for superheavy nuclei have been important parts of the experimental program. Separators were planned and built with the aim of identifying superheavy nuclei produced both in fusion as well as in transfer reactions. Mechanical transport systems and chemical separation techniques have been improved. (An overview can be found in ref. 2.) Although the predicted spherical superheavy nuclei have not been found yet, the sensitivity of the experiments can be increased, and nuclei produced with cross-sections as small as 3 pb identified.

In Darmstadt, isotopes of the new elements 107 (nielsbohrium, Ns), 108 (hassium, Hs) and 109 (meitnerium, Mt) have been identified from only a small number of decaying atoms produced with cross-sections of 167 pb [3], 19 pb [4] and 10 pb [5] respectively. The lowest cross-section measured up to now is $(3.2^{+6.1}_{-2.6})$ pb for the production of the isotope ^{264}Hs in the reaction ^{207}Pb(^{58}Fe, 1n)^{264}Hs [6]. The high sensitivity achieved in the experiments results from the following: (a) a stable, high current beam from the UNILAC, (b) a highly efficient separation of the reaction products by the SHIP velocity filter [7]; (c) a sensitive detector system with the feasibility to correlate detector-implanted reaction products with their subsequent radioactive decay chains [8, 9].

Using averaged values of the experiment parameters of the experiments cited earlier, a beam time to detect with 95% probability one event produced in a reaction with a cross-section of 1 pb can be estimated to be 161 days. In that case, the average counting rate would be one event per 54 days. This value was calculated with an average projectile current of 170 pnA (1 particle nanoampere $=6.24\times10^{9}$ particles s^{-1}), a target thickness of 500 μg cm^{-2}, a separator efficiency of 30% and a detector efficiency of 72%. Also included are the accelerator and experiment efficiencies $\epsilon_{acc}\epsilon_{exp}=$ 65%, defined as the ratio of beam-on-target time to calendar time. It considers, for example, losses resulting from control reactions, changes of ion sources and replacement of target wheels. Although beam times of about 5 months are not completely unrealistic for exceptional experiments, such as the search for superheavy nuclei, such long beam times certainly cannot be carried out regularly to measure data which are produced on a level of 1 pb only.

The natural upper limit of the counting rate in a 1 pb experiment is reached in the case of a separation and detection efficiency of 100%. This limit is still a function of the beam current, which in turn is limited

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by the accelerator parameters and target properties. Presently, a realistic current limit is about 1 pμA for beams from calcium to nickel. With target thicknesses of 600 μg cm^{-2}, and in the case of Gaussian excitation functions with a full width at half-maximum (FWHM) excitation energy of 5 MeV and maximum cross-section 1 pb, the counting rate amounts to 0.9 events per day for reactions such as that of ^{62}Ni + ^{208}Pb.

In Section 3, we will discuss how close this upper counting rate limit can be approached with techniques being developed at present. In Section 2, experiments will be discussed that require a sensitivity of 1 pb.

2. Experiments that require a sensitivity of 1 pb

2.1. *Spherical superheavy nuclei*

Recently, the stability of heavy and superheavy nuclei has been investigated theoretically using refined models based on the Nilsson–Strutinsky approach [10–14]. Pairing and zero-point energies have been carefully adjusted, while Coulomb redistribution energies were found to be important and a deformation space up to $\lambda_{\max} = 10$ has been considered in some cases. The ground state microscopic shell corrections show two minima: one of about -6 MeV, centered around ${}^{272}_{162}110$ with maximum deformation $\beta_2 = 0.22$; the other of about -9 MeV around ${}^{292}_{178}114$, forming an island of spherical superheavy nuclei (Fig. 1(a)). The binding energies, and beta, alpha and fission half-lives were calculated. The known data could be reproduced with good accuracy. A rough sketch of the half-lives obtained is shown in Fig. 1(b). As a result, we can deduce that most nuclei that can be produced with stable projectiles and the available targets are predicted to be α emitters with half-lives between 1 μs and 1 s.

In 1983, superheavy nuclei were searched for in experiments sensitive to alpha or fission decays with half-lives down to 1 μs [15]. Cross-section limits of 200 pb were obtained when investigating the reaction

$$ {}^{48}\mathrm{Ca} + {}^{248}\mathrm{Cm} \longrightarrow {}^{296}116^{*} $$

The relatively high limits reached result from the fact that the total beam time had to be split to cover five different beam energies.

In a repetition of the ^{48}Ca + ^{248}Cm experiment, it may be possible to reach cross-section limits of 1 pb. However, at present, it is still uncertain if this limit would be low enough. Therefore, a safer strategy to prepare experiments for the investigation of superheavy nuclei is cross-section measurements using thorium, uranium and heavier targets. Systematic data beyond the presently known cross-sections for the production of $Z = 106$ (the heaviest element made until now using very asymmetric reactions) would then allow us to readjust the model parameters of the reaction theories to obtain more accurate estimates for the production cross-sections of spherical superheavy nuclei.

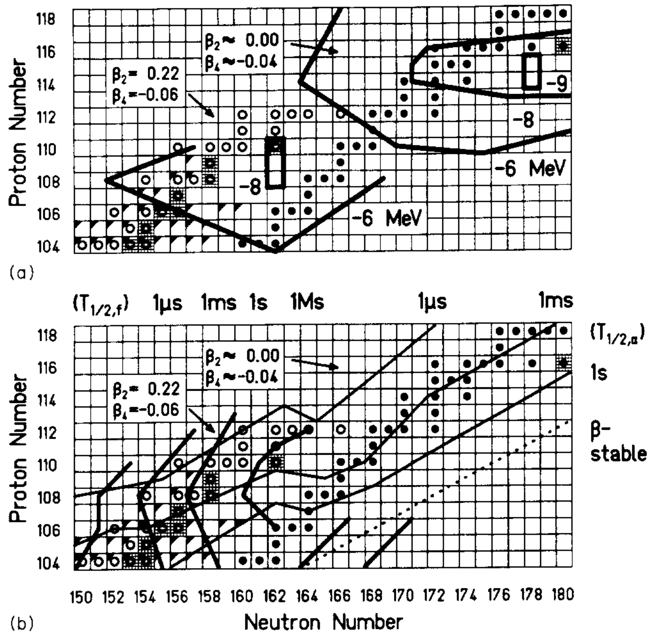

Fig. 1. Rough sketch of (a) calculated microscopic shell correction energies and (b) half-lives of even–even nuclei; bold lines, partial fission half-lives: fine lines, partial α half-lives; broken line, beta stability line. The data are from refs. 10, 11, 13, 14, 32 and 33. The fission half-lives of odd and odd–odd nuclei may be longer, as a result of additional hindrance factors. The arrows point to the region of strongly deformed nuclei centered around ^{272}110, and to the region of spherical superheavies at around ^{292}114; ○, compound nuclei of reactions (Ti to Zn) + ^{208}Pb → ($Z = 104$ to 112); ●, compound nuclei of reactions (O to Ti) + ^{248}Cm → ($Z = 104$ to 118); ▨, known nuclei; hatched area, compound systems investigated with SHIP. The figure does not include estimates for production cross-sections.

2.2. *The new elements 110 and 111*

In 1985 and 1986, two experiments were carried out at GSI in a search for element 110 [16, 17]. In both reactions, *i.e.* those of ^{64}Ni + ^{208}Pb and ^{40}Ar + ^{235}U, no event was observed that could be assigned to an isotope of element 110. The cross-section limits reached were 10 pb. These values are small enough to exclude experimentally an increase in the cross-sections compared with the systematics of lead-based reactions. Neither the addition of two neutrons to the $T_z = 3$ projectiles (^{50}Ti, ^{54}Cr, ^{58}Fe) used up to produce meitnerium nor the jump to the more asymmetric Ar + U reaction resulted in a cross-section increase by one or more orders of magnitude.

A fairly reliable cross-section estimate can be obtained for the reaction

$$ {}^{62}\mathrm{Ni} + {}^{208}\mathrm{Pb} \longrightarrow {}^{269}110 + 1\mathrm{n} $$

This reaction belongs to a series already known up to $Z=108$. Within the series, $T_z=3$ projectiles were always used for irradiation of ^{208}Pb targets. The cross-section systematics are shown in Fig. 2. The resulting cross-section estimate for the production of ^{269}110 is about 1 pb.

The predicted decay data of the isotope ^{269}110 are $E_\alpha \approx 11.3$ MeV, $T_{1/2}=10$–500 μs and $b_\alpha \approx 1$. Replacing the ^{208}Pb targets by ^{209}Bi leads to the compound nucleus ^{271}111. As a result of the odd–even effect for the cross-sections obvious in Fig. 2, an only slightly smaller cross-section (by a factor of 2 or 3) is expected for the 1n evaporation channel. In both cases, α decay will feed decay chains that are already well known from previous experiments. Delayed position and time coincidences will guarantee accurate identification of the isotopes produced.

2.3. Accurate excitation functions for production of elements up to Z = 108

The optimum projectile energy for the production of element 110 in the reaction of ^{62}Ni + ^{208}Pb cannot be chosen definitely on the bases of the present data. Figure 3 summarizes our knowledge on excitation energies for the production of elements 102–109 using lead- or bismuth-based reactions. From none of the reactions leading to the even elements 104, 106 and 108 has the maximum of the excitation function been measured.

The excitation functions for the production of heavy elements at projectile energies near the Coulomb barrier are very narrow (approximately 5 MeV FWHM) as a function of the excitation energy. Therefore, an accurate projectile energy has to be known to search for reaction products using recoil separators. In the case of the ^{62}Ni + ^{208}Pb experiment, this value can be estimated fairly reliably if the excitation functions of the reactions of ^{50}Ti + ^{208}Pb, ^{54}Cr + ^{208}Pb and ^{58}Fe + ^{208}Pb are known.

Reaction theories predict "extrapush" energies are necessary to fuse heavy nuclei. The amount of extrapush energy increases with increasing element number. This then results in higher excitation energies of the compound nuclei, leading to a decrease in fusion cross-sections, as a result of prompt fission. At present, the amount of extrapush energy required to obtain a maximum cross-section cannot be predicted accurately enough. However, one prediction [18] is shown in Fig. 3. An extrapush energy of nearly 30 MeV (center of mass system) has been calculated for the reaction of ^{62}Ni + ^{208}Pb.

Recently, Aguiar and coworkers [19, 20] included thermal fluctuations in one- and multidimensional trajectory models. Including fluctuations in these models makes it possible to predict the width of a trajectory ensemble leading to fusion or reseparation. The calculated widths are asymmetric and increase with the increasing average extrapush energies needed for compound nucleus formation. This is an important result for the production of heavy nuclei at low excitation energies, because fusion below the energy required to reach the unconditional saddle-point calculated with classical trajectories is no longer completely forbidden. Cross-section maxima are determined by the competition of complex fusion probabilities and prompt fission. The aim of experimental investigations is to measure the positions of the maxima and their values.

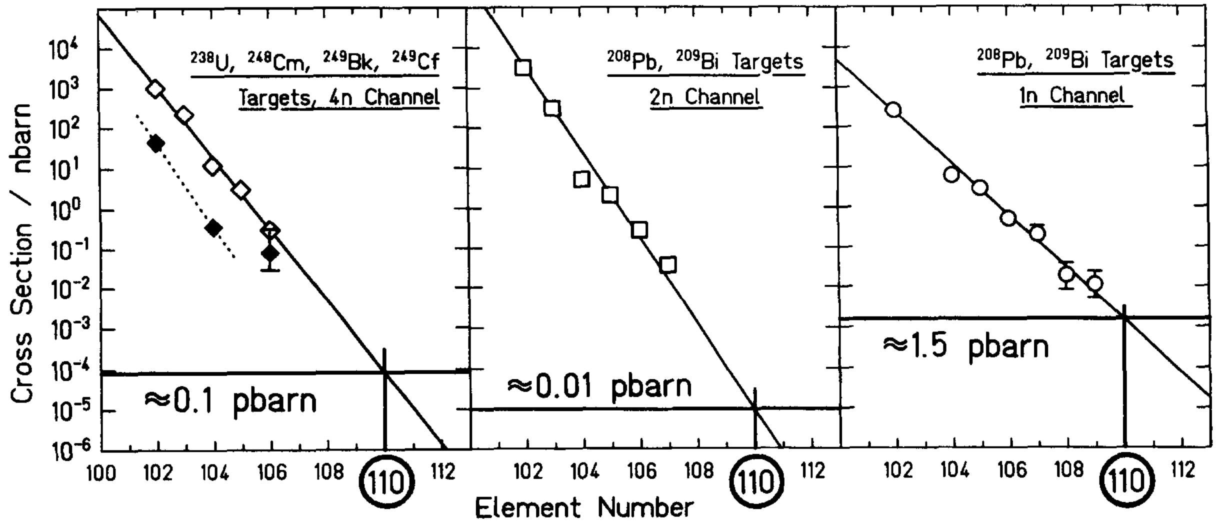

Fig. 2. Cross-section extrapolations up to element 110. The data are cross-sections of residues after fusion reactions and evaporation of one, two and four neutrons. Targets of ^{208}Pb, ^{209}Bi (Darmstadt data), ^{249}Bk, ^{249}Cf (Berkeley data) and ^{238}U, ^{248}Cm (full symbols, Dubna data) have been bombarded, respectively, with projectiles of equal or very similar isospin. The straight lines are least-squares fits to the data points. The favorite reaction for the production of element 110 is $^{62}\mathrm{Ni}+{}^{208}\mathrm{Pb}\rightarrow{}^{269}110+1\mathrm{n}$, with a cross-section of about 1 pb.

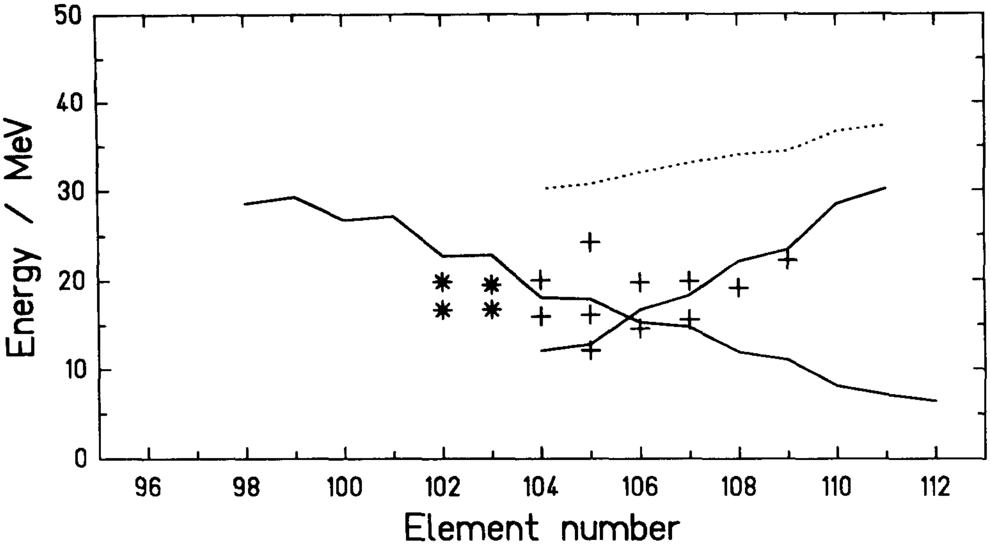

Fig. 3. Excitation energies at the Coulomb barrier for the production of heavy elements (descending curve) in reactions of $T_z = 3$ projectiles with targets of ^{208}Pb and ^{209}Bi respectively. The ascending curve shows an extrapush prediction [18]. Excitation energies at the barriers (including extrapush energies) are given by the broken line. The data points mark 1n channels observed in experiments using projectiles of the $T_z = 3$ series (+) and ^{48}Ca (*, $T_z = 4$). The excitation energies at the Coulomb barrier of the ^{48}Ca reactions are 17.7 MeV and 18.3 MeV in the cases of ^{256}102 and ^{257}103, respectively.

Using actinide targets, the heaviest element produced so far is $Z = 106$ [21–23]. A systematic investigation of the cross-sections for this type of reaction is necessary to prepare experiments for identifying neutron-rich, heavy element isotopes, especially spherical superheavy nuclei. The cross-section systematics shown in the left-hand part of Fig. 2 have still to be completed. Predictions based on a systematic investigation [24] of fusion reactions using actinide targets result in a value of 2 pb for the reaction ^{254}Es(^{20}Ne,4n)^{270}Mt.

2.4. Branching ratios

The stability of heavy nuclei is limited by fission, as a consequence of increasing Coulomb repulsion and decreasing shell effects. Therefore, the measured fission branching ratios and deduced partial fission half-lives of even–even nuclei are among the most interesting data to compare with theoretical investigations. Two examples are given below.

At present, the heaviest known, highest-Z, even–even nucleus is ^{264}Hs. The measured half-life of (76^{+364}_{-36}) μs agrees (within the error bars) with the calculated α half-lives. Different partial fission half-lives are predicted: 100 ms [11] and 10 μs [14]. The measured cross-section for ^{264}Hs was $(3.2^{+6.1}_{-2.6})$ pb. A lower experimental limit for the fission branching on a level of 1 pb and a more accurate half-life would help for a better understanding of the fission process, which depends sensitively on a complicated path in the potential energy surface and a deformation-dependent inertia.

The nucleus ^{258}106 is not yet known. Predicted α and fission half-lives are 2.7 ms [25] and 10 μs [11] respectively. The nucleus ^{258}106 is predicted to decay dominantly by fission, because α branching only accounts for 0.37%. For the production of ^{258}106, various combinations of chromium and lead isotopes can be used. Another possibility is a combination of vanadium plus ^{209}Bi. Therefore, an investigation of ^{258}106 will give interesting results on the stability of even–even heavy nuclei as well as on lead- and bismuth-based reaction processes. Another important result could be a correlation between low fission barriers and production cross-sections. Cross-sections are known for the production of ^{259}106 and ^{260}106 in 2n channels using as target ^{207}Pb and ^{208}Pb respectively. The values are about 300 pb. An investigation of the neighboring isotope ^{258}106 should be feasible with equipment sensitive down to 1 pb.

2.5. Level schemes: isomers, fine structure and nuclear deformations

Only little is known about excited levels of trans-fermium isotopes. Experimentally, excited levels can be studied after β and α decay, or by investigation of isomeric states. In all cases, the intensity is spread over a number of decay channels. For two reasons, high resolution is necessary: (1) the nuclei are strongly deformed and rotational levels are close in energy ($E_{2^+} \approx 40$ keV); (2) as a result of the calculated bunching of single-particle levels for neutron numbers $N = 152$ and 162 and proton number $Z = 108$, these nuclei have

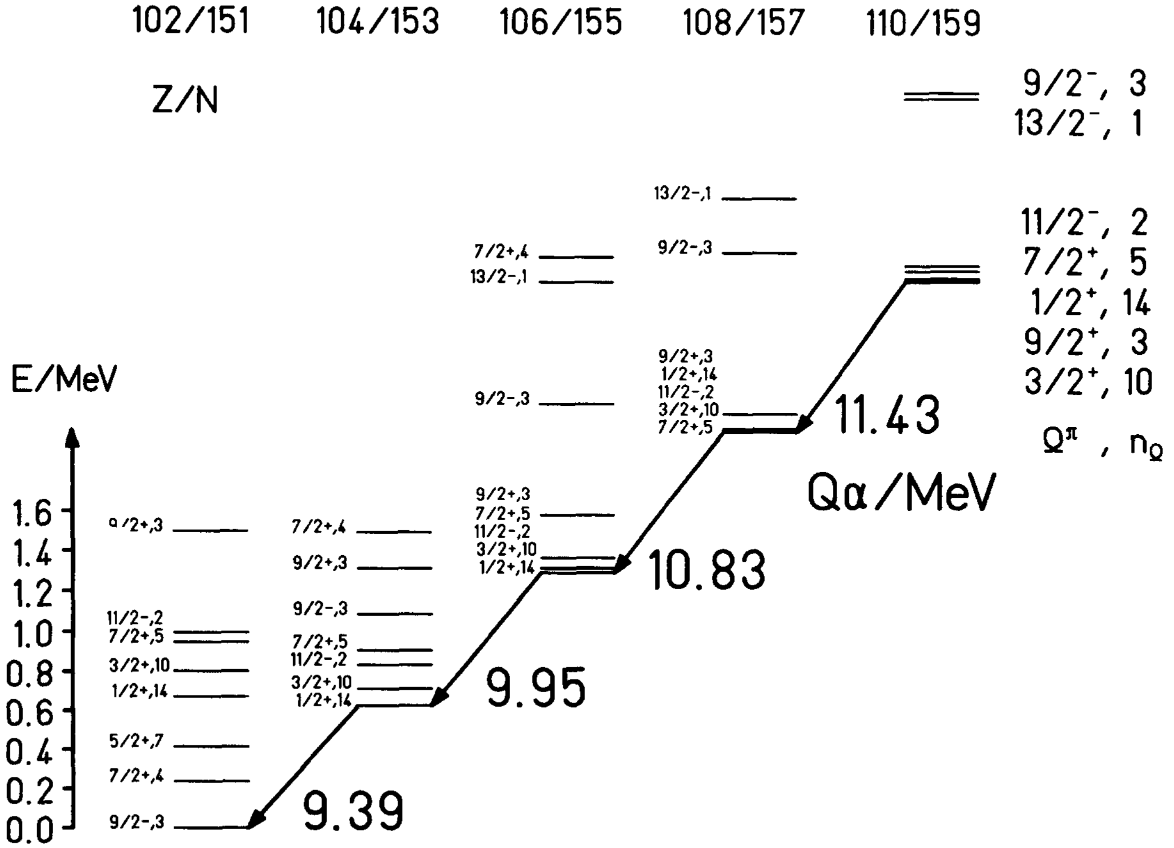

Fig. 4. Predicted α decay chain of $^{269}110$ [12].

a number of states with different K quantum numbers very close in energy. An example is shown in Fig. 4 for the predicted α decay chain beginning with $^{269}110$.

3. Technical developments to reach a sensitivity of 1 pb

Various types of reaction and identification technique have been applied in the past to investigate transuranium elements. The most successful methods for the production and identification of the heaviest elements have been fusion–evaporation reactions with heavy targets; recoil-separation techniques; and the identification of the nuclei by delayed coincidences to known daughter decays after implantation into position-sensitive detectors. This final technique has been further improved and is now most promising for the identification of new elements, the search for new isotopes and the measurement of new decay data of known nuclei.

3.1. *Recoil-separation techniques*

In contrast to the recoil-stopping methods, the recoil–separation techniques use the ionic charge and momentum of the recoiling fusion product obtained in the reaction process. Spatial separation from the projectiles and other reaction products is achieved by electric and magnetic fields. The separation times are determined by the recoil velocities and the lengths of the separators. Typically, a few microseconds are reached. Two types of recoil separator have been developed.

(a) The gas-filled separators use the different magnetic rigidities of the recoils and projectiles traversing a low pressure (about 1 mbar) gas-filled volume in a magnetic dipole field. Realizations are the small angle separator system (SASSY) at Berkeley [26], the helium charge exchange kaleidoscope (HECK) at Darmstadt [27, 28], and the gas-filled magnetic separator at Dubna [29].

(b) Wien-filter type of separators are based on the different electric rigidities of fusion products, projectiles and other reaction products after escape from solid state targets into vacuum. Ionic charge achromaticity is essential for high transmission. It can be achieved by additional magnetic fields or symmetric arrangements of electric fields. Realizations used in experiments for the investigation of heavy elements are the separator for heavy ion reaction products (SHIP) at Darmstadt [7] and the recoil energy separator VASSILISSA at Dubna [30].

3.2. Detectors

Recoil separators are designed to filter out with high transmission, nuclei that are produced in various evaporation channels. Because highly efficient separation cannot be achieved background free, the transmitted particles have to be identified by detector systems. The detector type to be selected depends on the particle rate, energy, decay mode and half-life. Experimental as well as theoretical data on the stability of heavy nuclei show that they decay by α emission or fission, with half-lives ranging from microseconds to days. Therefore, silicon semiconductor detectors are most suitable for the identification of nuclei and measurement of their decay properties.

If the total rate of separated ions is low, the particles can be implanted directly into the silicon detectors. Using position-sensitive detectors, the local distribution of the implanted particles is measured. In that case, the detectors act as diagnostic elements to optimize and control the ion optical properties of the separator.

If the implanted nuclei are radioactive, then the same position is measured from the decay process as obtained from the decays of the daughter products. This is the case because the recoil effects are small compared with the range of implanted nuclei, emitted α particles or fission products, and compared with the detector resolution. Recording the data event by event allows us to analyse delayed coincidences with variable position and time windows for identification of the decay chains.

This method was first used for the identification of neutron-deficient α emitters and to search for proton radioactivity near $N=82$ [8]. The detector system was enlarged and an array of seven position-sensitive silicon detectors [9] was used to identify the elements 107, 108 and 109. The same method was used at Berkeley in an experiment to search for superheavy nuclei [15]. A further enlarged system has been build at Darmstadt for a planned experiment to search for element 110, and for further investigation of the decay properties of lighter nuclei [31] (Fig. 5). It is composed of seven identical 16-strip silicon wafers plus a germanium detector.

In front of the system, two secondary electron foil detectors are mounted 500 mm apart from each other. Their signals are used to distinguish implantations from radioactive decays of implanted nuclei. Because of the high efficiency of these detectors (99% or more), very clean decay spectra are obtained, and the coincidence time for measuring decay chains is significantly prolonged. The time resolution is about 500 ps, which is small enough to obtain, together with the energy signals from the silicon detector, a rough mass assignment for the implanted ions.

4. Conclusions

Experimental techniques, based on previous successful experiments on the investigation of heavy element production and stability, have been further developed. Experimental search techniques for new heavy nuclei or rare decay modes produced with cross-sections

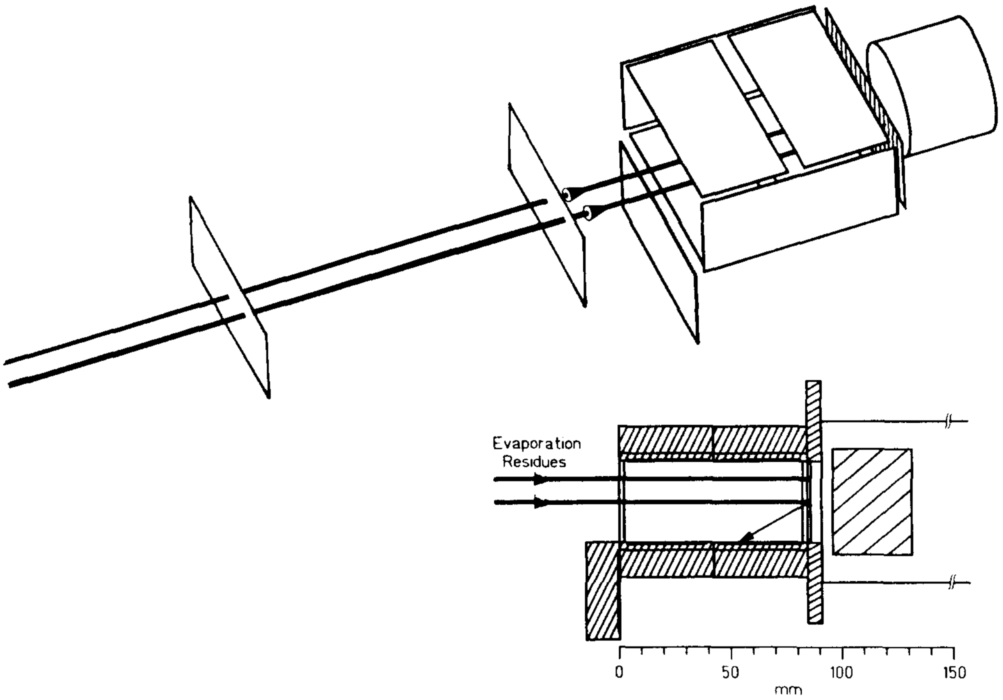

Fig. 5. New assembly of position-sensitive silicon strip detectors.

of picobarn and even somewhat below are within reach. Not unrealistic estimates for the rate of separated heavy nuclei in reactions with ^{208}Pb targets at cross-sections of 1 pb result in a value of 0.4 per day. A beam current of 6×10^{12} ions s^{-1}, a target 600 μg cm^{-2} thick, a separator transmission of 50% and a detection efficiency of 100% have been assumed. Repetition of the search experiments for superheavy nuclei could lower the presently existing cross-section limits by more than two orders of magnitude. In the case of lighter elements, which are produced with higher cross-sections, the search for α decay fine structure, isomeric states, and small α, fission or β branching ratios are the aim of planned experiments.

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