

# Beyond darmstadtium — Status and perspectives of superheavy element research

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**Abstract.** The search for superheavy elements has yielded exciting results for both the “cold fusion” approach with reactions employing Pb and Bi targets and the “hot fusion” reactions with  $^{48}\text{Ca}$  beams on actinide targets. In recent years the accelerator laboratories in Berkeley, Dubna and Darmstadt have been joined by new players in the game in France with GANIL, Caen, and in Japan with RIKEN, Tokyo. The latter yielding very encouraging results for the reactions on Pb/Bi targets which confirmed the data obtained at GSI. Beyond the successful synthesis, interesting features of the structure of the very heavy nuclei like the hint for a possible K-isomer in  $^{270}\text{Ds}$  or the population of states at a spin of up to  $22\hbar$  in  $^{254}\text{No}$  give a flavor of the exciting physics we can expect in the region at the very extreme upper right of the nuclear chart. To get a hand on it, a considerable increase in sensitivity is demanded from future experimental set-ups. High intensity stable beam accelerators, mass measurement in ion traps and mass spectrometers, as well as the possible employment of unstable neutron-rich projectile species, initially certainly only for systematic studies of reaction mechanism and nuclear structure features for lighter exotic neutron-rich isotopes, are some of the technological challenges which have been taken on.

**PACS.** 24.75.+i General properties of fission – 25.70.Gh Compound nucleus – 25.70.Jj Fusion and fusion-fission reactions

## 1 Introduction

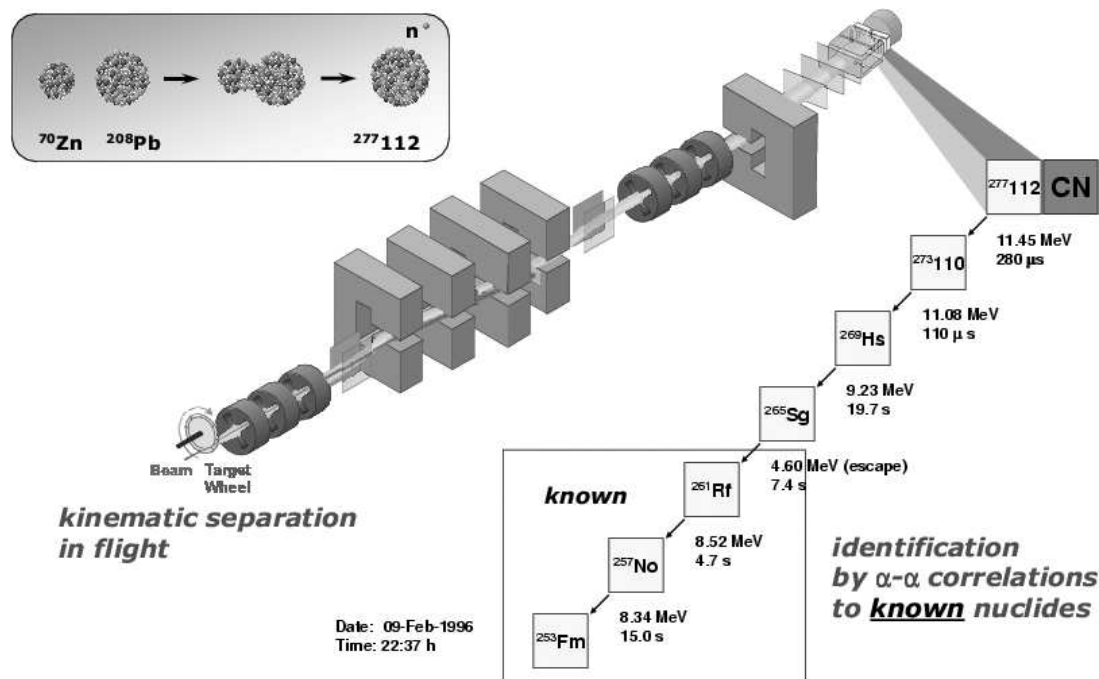
The community engaged in the synthesis and investigation of superheavy elements (SHE), traditionally undertaken at the FLNR/JINR in Dubna, Russia at the LBNL in Berkeley, California (USA) and at GSI in Darmstadt, Germany has recently been joined by other laboratories. In particular GANIL in Caen, France and RIKEN in Tokyo, Japan have started substantial experimental programs in this field of nuclear physics. Combined efforts have yielded impressive results in both approaches the hot fusion with actinide target, recently performed successfully using  $^{48}\text{Ca}$  beams at Dubna [1], and the fusion on Pb and Bi targets leading to compound systems of relatively low excitation energy. Whereas the Dubna results still lack confirmation, despite various attempts at LBNL for the reaction  $^{48}\text{Ca} + ^{238}\text{U}$ , the groups at GANIL and, in particular, at RIKEN succeeded in reproducing the GSI results. Here an overview over the recent development at LBNL, GANIL and RIKEN will be given after a more detailed description of the present activities at GSI including the achievements in the synthesis of heavy elements and the investigation of the nuclear structure of heavy isotopes.

The present sensitivity limit is at a production cross section of  $\approx 1$  pb. The efforts to push this limit down to even lower values at GSI by various development activities to increase the set-up sensitivity are presented at the end of the paper.

## 2 Synthesis and identification of superheavy elements at GSI/SHIP

The identification of superheavy elements in heavy ion fusion reactions is based on two major ingredients: the separation of the fusion products in flight from the beam particles and the identification of the products via evaporation residue(ER)- $\alpha$  correlations. The velocity filter SHIP at GSI provides separation, using the velocity difference between the faster beam and the slower fusion products in the fashion of a classical Wien-filter, via the comparison of crossed  $E$ - and  $B$ -fields —in the case of SHIP in a separated field configuration. The particles passing the velocity filter are then implanted into a position sensitive silicon strip detector set-up where position, time and energy of the fusion products and subsequent decays by  $\alpha$  emission and spontaneous fission are recorded. The  $Z$  and  $A$  identification of the starting point of those decay chains is unambiguously provided by the connection to known  $\alpha$  emitters

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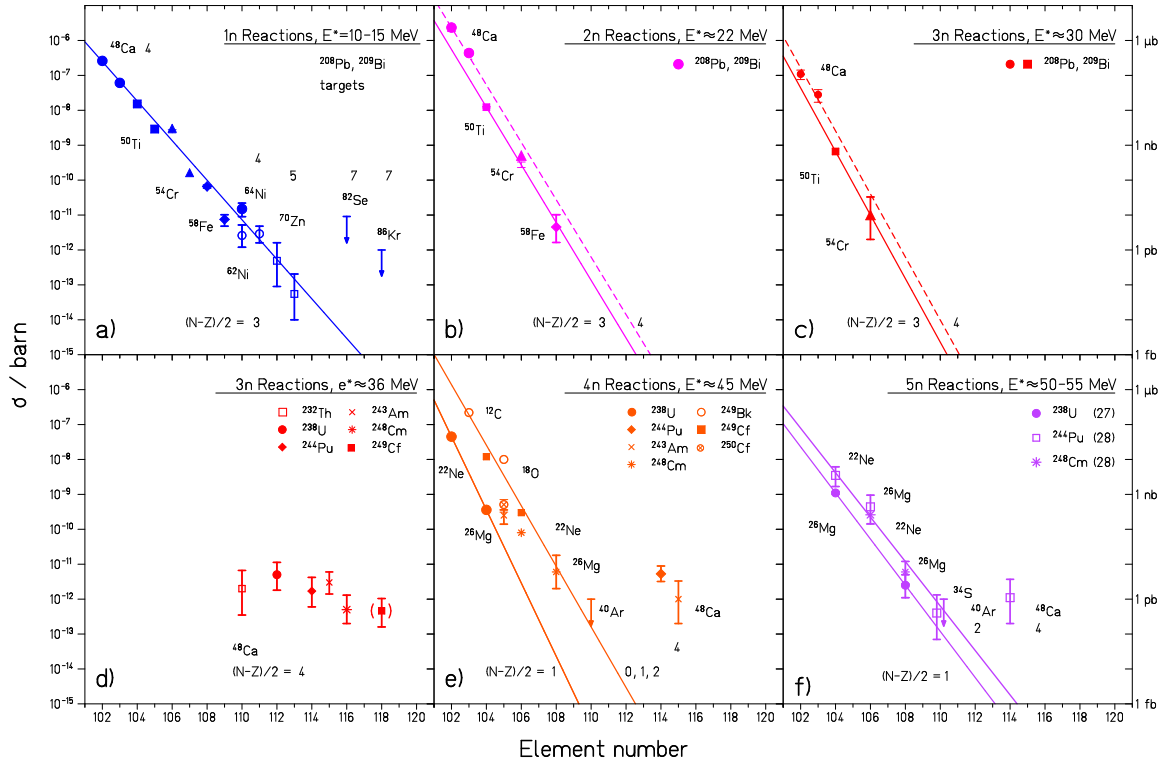


**Fig. 1.** Separation, detection and identification of fusion reaction products with the velocity filter SHIP and the ER- $\alpha(-\alpha)$  correlation method. As an example the first decay chain observed in the reaction  $^{70}\text{Zn} + ^{208}\text{Pb} \rightarrow ^{277}_{112} + 1n$ .

at the end of the decay sequence. In fig. 1 the method is illustrated for the example of the first decay chain, observed in the reaction  $^{70}\text{Zn} + ^{208}\text{Pb} \rightarrow ^{277}_{112} + 1n$  in 1996 ref. [2].

The elements with  $Z = 107$ –112 have been synthesized and unambiguously identified at SHIP. The elements 107–111 have already been named and have been entered as bohrium (Bh,  $Z = 107$ ), hassium (Hs,  $Z = 108$ ), meitnerium (Mt,  $Z = 109$ ), darmstadtium (Ds,  $Z = 110$ ) and roentgenium (Rg,  $Z = 111$ ) in the periodic table of elements. A summary of these experiments can be found in ref. [3]. The heaviest nucleus synthesized so far at SHIP is the isotope  $^{277}_{112}$ . In early 1996 the search for element 112 was undertaken using the projectile target combination  $^{70}\text{Zn} + ^{208}\text{Pb}$ . An excitation energy of  $E^* = 10.1$  MeV was chosen. One decay chain which could be attributed to  $^{277}_{112}$  was observed. The resulting production cross section was  $\sigma = (0.37^{+0.85}_{-0.31})$  pb. In a recent experiment in May 2000 a second decay chain of  $^{277}_{112}$  has been recorded. This latter chain has been observed at an excitation energy of about 2 MeV higher at  $E^* = 12$  MeV. During an irradiation time of 19 days a total of  $3.5 \times 10^{18}$  projectiles were sent onto the target. The resulting cross section at this energy is  $(0.49^{+1.12}_{-0.40})$  pb. This value fits well into the cross section systematics for 1n reaction channels shown in fig. 2 upper left panel. The first two  $\alpha$ -decays have energies of 11.17 and 11.20 MeV, respectively. They are succeeded by the emission of an  $\alpha$ -particle that exhibits with 9.18 MeV a decay energy lower by 2 MeV. Correspondingly, the lifetime increases by about five orders of magnitude between the second and third  $\alpha$ -decay. This decay pattern is in agreement with the one observed for the chain in the first experiment and supports the explanation of

a local minimum of the shell correction energy at neutron number  $N = 162$ , which is crossed by the  $\alpha$ -decay of  $^{273}\text{Ds}$ . The  $\alpha$  energy of 9.18 MeV for  $^{269}\text{Hs}$  is within the detector resolution identical to the one observed in the first chain. A new result is the occurrence of fission ending the new chain at  $^{261}\text{Rf}$ , for which fission was not observed so far, but is likely to occur taking into account the high fission probabilities of the neighboring isotopes. For more details see ref. [2]. An experiment performed at GSI in April/May 2001 and designed to investigate the chemical properties of Hs confirmed these findings. Ch.E. Düllmann *et al.* [4], employing the reaction  $^{26}\text{Mg} + ^{248}\text{Cm}$ , observed in the decay chain of the 5n-evaporation channel which enters our  $^{277}_{112}$  decay sequence at  $^{269}\text{Hs}$  both, a fission and an  $\alpha$ -decay branch. Finally K. Morita *et al.* [5] performed the same reaction as employed at SHIP. They observed two decay chains assigned to  $^{277}_{112}$  where they observed in both cases fission of  $^{261}\text{Rf}$ , confirming the GSI results. In fig. 2 the cross section systematics for both approaches, fusion on  $^{208}\text{Pb}$  and  $^{209}\text{Bi}$  (“cold fusion”; upper panels) and on actinide targets (“hot fusion”; lower panels) are compared, for 1n-3n and 4n-5n evaporation reaction channels, respectively [6]. Whereas for the “cold fusion” reactions no deviation from the steep decrease of the maximum cross section with increasing  $Z$  is observed, the Dubna results for the “hot fusion” show cross-section values which surprisingly remain rather high and constant around and above the 1-pb-level for  $Z$  up to 116(118) [1]. Despite a by now relatively vast body of data accumulated and assigned to nuclei in the region of  $Z = 112$ –116, the  $Z$  and  $A$  assignments are still not firm as in those cases the decay chains are not connected to known  $\alpha$  emitters, but end in unknown fissioning nuclei.



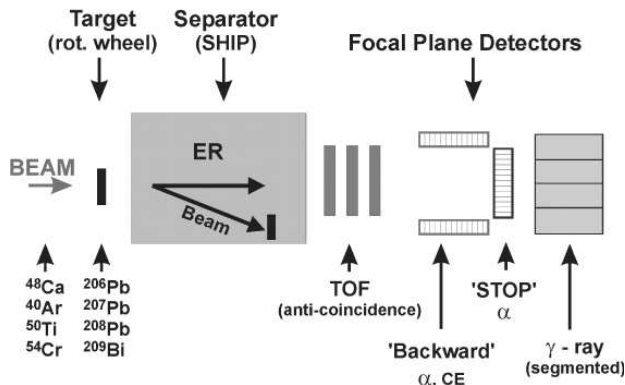
**Fig. 2.** Maximum cross section systematics for reactions with Pb- and Bi-targets (upper panels; 1n-3n reaction channels) and for actinide targets (lower panels; 3n-5n reaction channels) [6].

## 2.1 Nuclear structure investigations of heavy nuclei at SHIP

The detailed understanding of nuclear structure and its development in the vicinity of closed shells, in regions of deformation and towards heavier masses and higher  $Z$  is a necessary ingredient for a successful progress in the synthesis of new heavy elements. The possible trends in single particle levels are the most sensitive probe for the formation of low level density, and eventually the appearance of shell gaps and regions of (shell-) stabilized nuclei. Decay spectroscopy of  $\alpha$ -emitters stopped after separation is a powerful tool to study their daughter products or isomeric states via  $\alpha$  fine structure or  $\alpha$ - $\gamma$  spectroscopy by ER- $\alpha$  or ER- $\alpha$ - $\gamma$  coincidence measurements. Here the fusion reaction products are after separation implanted into a solid state (“stop”) detector for the residue and  $\alpha$  detection which is combined with a high resolving  $\gamma$ -ray (Ge-) detector as shown in fig. 3. This method is very clean as compared to in-beam studies because of the effective shielding from target background due to its spacial separation and the effective cleaning by the ER- $\alpha$  coincidence technique. It is highly efficient because of the favorable close geometry of the  $\alpha$  and  $\gamma$  detectors and the very well localized stopped  $\gamma$ -ray source the implantation spot of the ER is forming on the stop detector. The latter has the further advantage of the absence of any  $\gamma$ -ray Doppler shift or broadening which yields, with a moderate crystal size and

a moderate granularity of the Ge-detector, a nevertheless high efficiency  $\epsilon$ . In case of the SHIP set-up we achieve an  $\epsilon$  of up to  $\approx 15\%$ . We have applied the technique of  $\alpha$  fine structure and  $\alpha$ -( $\alpha$ )- $\gamma$  spectroscopy to study several radium isotopes ( $A = 209-212$ ) [7], neutron-deficient nuclei with  $Z = 86-92$  [8] up to the isotopes  $^{252,253}\text{Lr}$ ,  $^{255}\text{Rf}$  and  $^{256,257}\text{Db}$  [9]. In fig. 4 as an example the  $\alpha$ - $\gamma$  coincidence spectra for the latter case and the conclusions one can draw in terms of level schemes and single particle level systematics are shown.

The nuclear structure of heavy nuclei with  $Z \geq 82$  is interesting in itself as many interesting features, like *e.g.* isomers, shape transitions and shape co-existence, are expected and found in this region. One recent example is the hint for a K-isomer in  $^{270}\text{Ds}$  we reported recently [10]. In an experiment in October 2000 we observed in the reaction  $^{64}\text{Ni} + ^{207}\text{Pb}$  eight decay chains of correlated ER- $\alpha$ -fission events which we attribute to the decay of the new isotope  $^{270}\text{Ds}$ . Also the daughter and grand daughter products  $^{266}\text{Hs}$  and  $^{262}\text{Sg}$  had not been observed before. Here the production cross section remained surprisingly high as compared to the more neutron rich  $^{271}\text{Ds}$  at  $13 \pm 5$  pb produced in the reaction  $^{64}\text{Ni} + ^{208}\text{Pb}$ . Moreover, from the measured decay data hints for interesting nuclear structure properties could be deduced. The observed eight decay chains could be attributed to two different half-lives, 0.15 ms and 8.6 ms, respectively. Together with the systematics of  $\alpha$ -decay energies and a 218 keV  $\gamma$ -ray



**Fig. 3.** Schematic view of a decay spectroscopy set-up consisting of a separation stage (SHIP) and a  $\alpha$ - $\gamma$  coincidence detector arrangement. In addition transmission detectors for time of flight and veto purposes are shown. At SHIP reactions like  $^{48}\text{Ca}$ ,  $^{40}\text{Ar}$ ,  $^{50}\text{Ti}$ ,  $^{54}\text{Cr}$  +  $^{206,207,208}\text{Pb}$ ,  $^{209}\text{Bi}$  are investigated.

measured in coincidence to one of the emitted  $\alpha$ -particles, this was interpreted as the simultaneous population of the ground state and a K-isomer. Support for this interpretation was provided by theoretical calculations which predict spin and parity values of  $8^+$ ,  $9^-$  or  $10^-$  [11]. Recent calculations suggest the occurrence of K-isomers is a general feature in the region of heavy nuclei [12].

## 2.2 Reaction mechanism studies

Complete fusion reactions appear to be the most successful method for the production of transactinide nuclei. The formation cross section of a specific nuclide in a given reaction, however, is strongly dependent on the excitation energy  $E^*$  of the compound nucleus, according to the relation  $E^* = E_{\text{cm}} + Q$  (where  $E_{\text{cm}}$  denotes the energy in the center-of-mass system and  $Q$  the  $Q$ -value of the reaction), and thus on the bombarding energy  $E_{\text{lab}} = (m_p + m_t)/m_t \times E_{\text{cm}}$  (where  $m_p$  and  $m_t$  denote the masses of projectile (p) and target (t), respectively). Since maximum production cross sections are decreasing rapidly with increasing atomic numbers, the choice of the optimum  $E_{\text{lab}}$  is crucial for the production of the heaviest nuclei. It is, together with the understanding of the nuclear structure of the very heavy nuclei crucial as input knowledge for a successful program to extend the synthesis of new elements to higher  $Z$  and eventually to the region of the spherical superheavy nuclei.

## 3 Synthesis and identification of superheavy elements in other laboratories

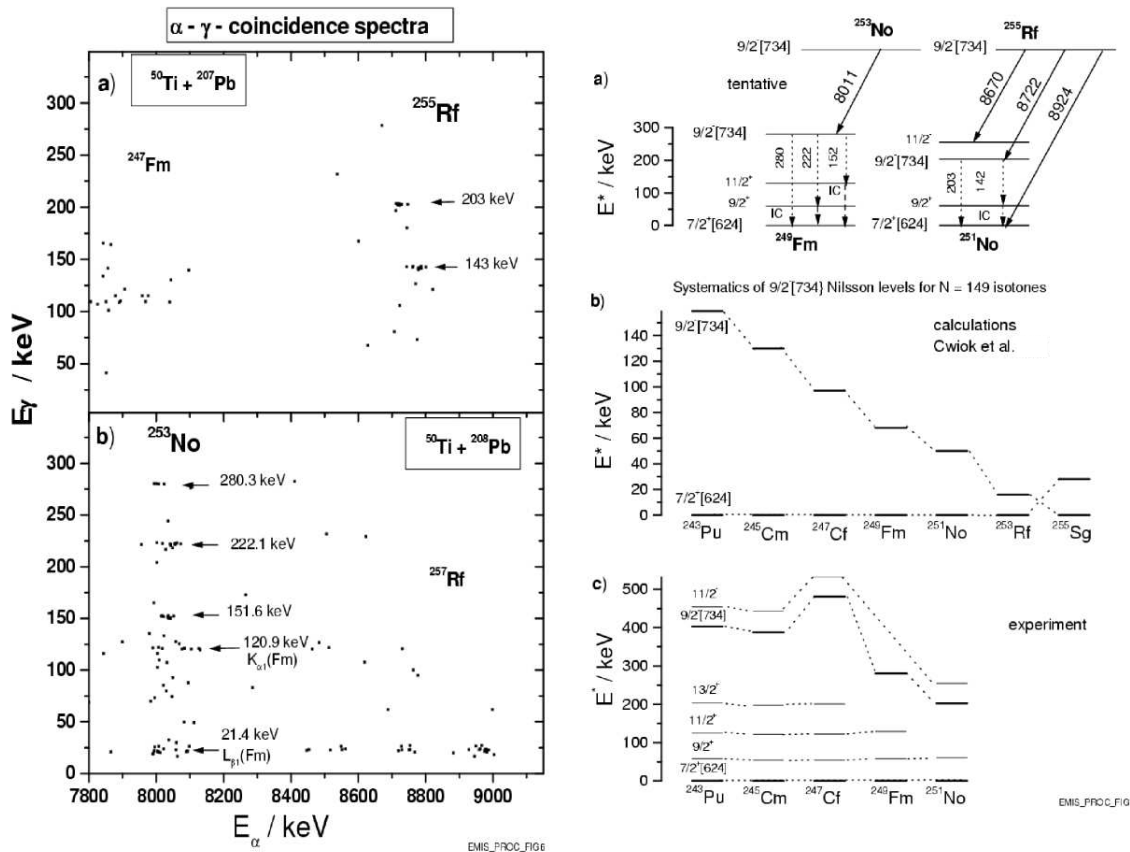
The recent results for the  $^{48}\text{Ca}$  induced reactions obtained at the FLNR/JINR in Dubna are presented in detail in ref. [1]. I shall here give a brief review on the recent achievements at the LBNL in Berkeley CA, USA, GANIL in Caen, France and RIKEN in Tokyo, Japan. All three laboratories use as at the FLNR and in contrast to GSI

a 100% duty cycle accelerator. At Berkeley with the BGS and at RIKEN with GARIS a gas-filled separator is the heart of the SHE-production set-up, whereas at GANIL this part is taken by the velocity filter of LISE.

At the BGS in Berkeley both approaches for SHE synthesis, cold and hot fusion are followed [13,14]. They investigated, *e.g.*, the excitation function for the reaction  $^{64}\text{Ni} + ^{208}\text{Pb}$  and compared their results for  $^{271}\text{Ds}$ , the 1n reaction channel, with the ones obtained at GSI and RIKEN. The maxima for the three data sets are shifted by  $\approx 2$  MeV between each other with the GSI results being the lowest, the RIKEN ones the highest and the LBNL data in between. This observation, however, has to be put in relation to the energy loss in the target of  $\approx 2.5$  MeV to 6.5 MeV and the uncertainty of the cross section values due to the low statistics. Another interesting result obtained at the BGS is a cross section for  $^{272}\text{Rg}$  of  $\sigma = 1.7^{+3.9}_{-1.4}$  pb observed in the reaction  $^{65}\text{Cu} + ^{208}\text{Pb}$  which is within error bars comparable to the one observed at GSI for the same isotope in the reaction  $^{64}\text{Ni} + ^{209}\text{Bi}$  with  $\sigma = 3.5^{+4.4}_{-2.3}$  pb. This observation suggests the use of odd- $Z$  projectiles on  $^{208}\text{Pb}$  in other cases like *e.g.*  $^{55}\text{Mn} + ^{208}\text{Pb}$  where an excitation function has been measured. As far as the hot fusion approach is concerned, an attempt to reproduce the Dubna findings for  $^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{283}112 + 3n$  remained unsuccessful with a cross section limit of  $\sigma_{\text{limit}} < 1$  pb.

At GANIL as a first step to enter the field of SHE research the excitation functions for seaborgium and hassium isotopes were measured using the same reactions as in the SHIP experiments,  $^{54}\text{Cr} + ^{208}\text{Pb}$  and  $^{58}\text{Fe} + ^{208}\text{Pb}$  [15]. The obtained results are in good agreement with the GSI data. From the comparison of the yields a relatively low transmission of  $\approx 15\%$ – $17\%$  of the LISE velocity filter was deduced. In November 2004 the reaction  $^{76}\text{Ge} + ^{208}\text{Pb}$  was employed to search for  $^{283}114$ . At a beam energy of  $E_{\text{beam}} = 5.02$  AMeV with an intensity of  $> 1$  particle  $\mu\text{A}$  a beam dose of  $5 \times 10^{18}$   $^{76}\text{Ge}$  projectiles was put onto the  $420 \text{ mg/cm}^2$  thick targets. This corresponds to a sensitivity of 0.6 pb if one event would have been observed. However, no event could be attributed to  $^{283}114$ . This results in a cross section limit of  $\sigma_{\text{limit}} = 1.2$  pb in the center-of-mass energy range 274.5 MeV–278.5 MeV.

Also at the gas-filled separator GARIS of RIKEN the first steps had been made reproducing GSI results of reactions with Pb and Bi targets. The series of successful experiments was started with the observation of 10 decay chains of  $^{265}\text{Hs}$ . For both isotopes  $^{271}\text{Ds}$  and  $^{272}\text{Rg}$  14 decay chains were produced [16] before Morita and co-workers succeeded in the reproduction of  $^{277}112$ . They obtained two decay chains, each terminating with the fission of  $^{261}\text{Rf}$  and thus confirming the SHIP results [5]. Finally in two long runs of about 140 days of  $^{70}\text{Zn}$  beam on a  $^{209}\text{Bi}$  target they observed on July 23<sup>rd</sup> one decay chain for the new isotope  $^{278}113$  with the extremely low production cross section of  $\sigma = 55^{+154}_{-47}$  fbarn [17]. The observed decay chain ended with  $\alpha$ -decay and fission of the known isotopes  $^{266}\text{Bh}$  and  $^{262}\text{Db}$ , respectively.



**Fig. 4.** Left panels:  $\alpha$ - $\gamma$ -coincidence spectra for the reactions  $^{50}\text{Ti} + ^{208}\text{Pb}$  (a) and  $^{50}\text{Ti} + ^{207}\text{Pb}$  (b). Right panels: tentative level schemes for  $^{253}\text{No} \rightarrow ^{249}\text{Fm}$  and  $^{255}\text{Rf} \rightarrow ^{251}\text{No}$  (a), calculated (b) and measured (c) trend for first excited states for a series of  $N = 149$  isotones [9].

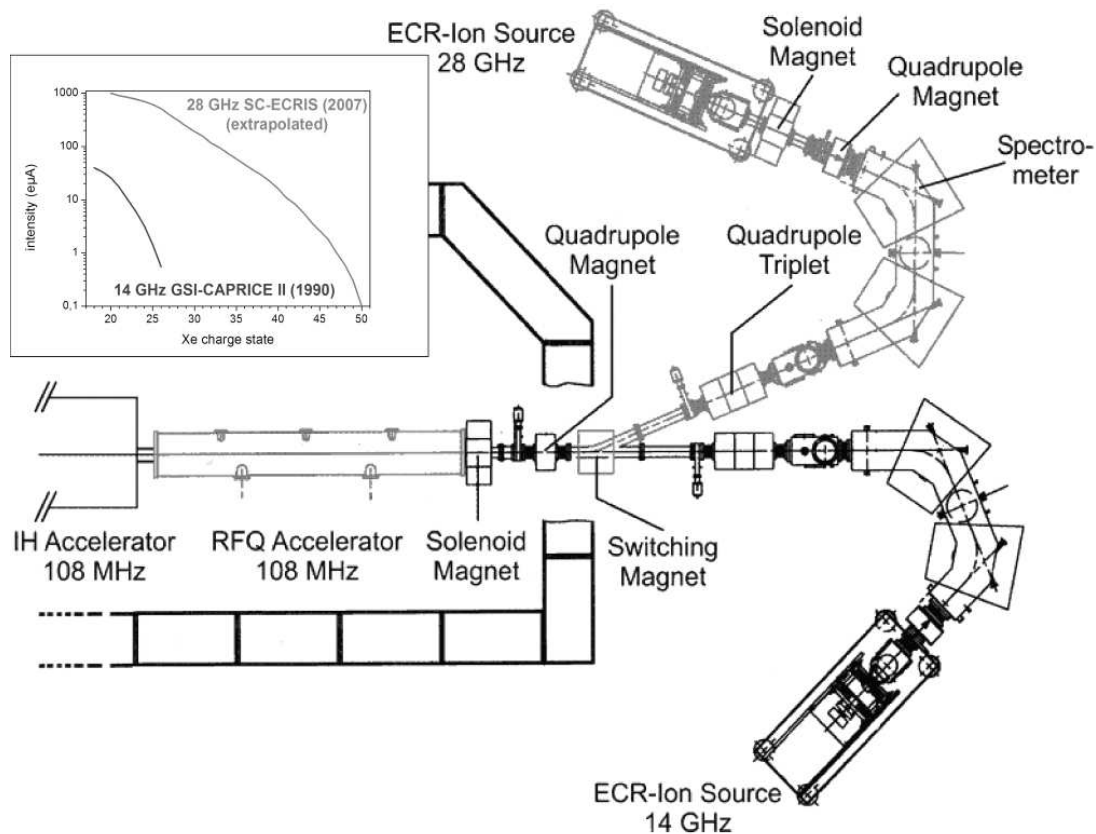
## 4 Technical development at GSI/SHIP

To access a region of lower cross section the number of interactions and, therefore, the number of projectiles has to be increased. The UNILAC at GSI delivers the beam with a duty cycle of about  $\leq 28\%$ . Apart from raising the beam current, the use of an accelerator with 100% duty cycle (DC) would provide a factor of 3.5 higher in beam intensity. Such a CW-linac is presently being studied by the group of Ratzinger *et al.* at the University of Frankfurt, Germany. As a first step an accelerator upgrade comprising a 28 GHz superconducting ECR ion source together with an adapted RFQ injection accelerator structure is presently being proposed. This configuration will via a substantial increase of charge state and injection beam intensity yield an increase in beam intensity of a factor  $\approx 10$  as compared to the present UNILAC beam intensities. Figure 5 shows the new ion source combined with the existing 14 GHz normal conducting ERC source. The insert shows a comparison for the performance of both sources in terms of the extracted beam intensity as a function of the charge state for Xe-ions. The higher charge state achievable with the 28 GHz SC-ECRIS source will allow for a UNILAC duty cycle of  $\approx 50\%$ . The increased beam current asks for measures to protect the Pb and Bi targets,

both having low melting points at 600.6 K and 544.5 K, respectively. Chemical compounds of Pb or Bi with higher melting temperatures have been successfully tested. The metallic targets have now been replaced by PbS and Bi<sub>2</sub>O<sub>3</sub> with melting points at 1400 K and 1090 K, respectively. In addition the possibility of an active target cooling has been investigated and will allow for an additional increase of heat power to deposited in the target [18].

## 5 Open tasks

The localization of the region of spherical shell-stabilized superheavy nuclei is still an open task. The results from the FLNR, Dubna seem to have come closer to this region. The non-connected decay chains, however, are at the moment an important and demanding challenge that requires still substantial effort in order to obtain a firm isotopical assignment. At SHIP the investigation of the reaction  $^{48}\text{Ca} + ^{238}\text{U}$  as a first approach to study this region is planned for spring/summer 2005. Additional information like the mass of the reaction products will help for a more reliable positioning of observed decay patterns in  $A$ , and possibly  $Z$ . Technical development in this direction is presently ongoing at GSI and with SHIPTRAP one promising tool is



**Fig. 5.** Upgrade of the UNILAC high charge state injector by a new 28 GHz super-conducting ECR ion source and a new RFQ pre-acceleration structure. Insert: comparison of the new 28 GHz ECR ion source with the present 14 GHz ECR source in intensity (vertical axis) and charge state (horizontal axis).

presently being put into operation [19]. Simultaneously systematic investigations of the structure of very heavy nuclei and the reaction mechanism governing the process of fusion and survival against fission are pursued. Together with the already achieved results this will yield valid input data for the more and more sophisticated models. With an increasing understanding of the properties of the very heavy nuclei one can hope to better localize the region of the spherical superheavy nuclei and to favor the experimental approach to it. High currents of stable beams and radioactive beams are options for the future.

The recent experiments were performed together with H.-G. Burkhard, F.P. Heßberger, S. Hofmann, B. Kindler, I. Kojouharov, P. Kuusiniemi, R. Mann, G. Münzenberg, B. Lommel, H.-J. Schött, J. Steiner, B. Sulignano (GSI Darmstadt), A.N. Andreyev, A.G. Popeko, A.V. Yeremin (FLNR-JINR Dubna), S. Antalic, P. Cagarda, Š. Šaro, B. Streicher (University of Bratislava), J. Uusitalo and M. Leino (University of Jyväskylä).

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