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Chemistry

TASCA

Theory

Observation of ^{270}Hs in the complete fusion reaction ^{36}S+^{238}U*

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Superheavy elements ($Z \geq 104$) exist only due to nuclear shell effects. Deformed shell closures at Z=108 and at N=162 have been predicted theoretically [1] and observed in recent experiments [2]. Three hassium isotopes $^{269\text{-}271}$Hs, have been produced in the complete fusion reaction ^{248}Cm(^{26}Mg, xn)$^{274\text{-}x}$Hs at the linear accelerator UNILAC at GSI [2,3]. The excitation function measurement at five different beam energies resulted in maximum cross sections of the 3n, 4n, 5n exit channels of a few pb [3].

Recent theoretical studies, analysing the formation of ^{270}Hs in the 4n channel, using a two-parameter Smoluchowski equation, predict even higher cross sections due to a lower reaction Q-value for the reaction ^{238}U(^{36}S,4n)^{270}Hs (24 pb) and ^{226}Ra(^{48}Ca,4n)^{270}Hs (30 pb) [4]. HIVAP [5] predicts a maximum cross section of 3.5 pb for the reaction ^{238}U(^{36}S,4n)^{270}Hs.

Here, we report on first results of an experiment aimed to study the nuclear fusion reaction ^{36}S + ^{238}U, leading to the compound nucleus ^{274}Hs*. For the experiment we used our highly efficient chemical separation and detection system COMPACT which was connected to a Recoil Chamber (RC) installed behind the ARTESIA target wheel in cave X1 [2]. A beam of $^{36}S^{5+}$ ions was accelerated by the UNILAC up to 7.13 MeV/u and impinged on a rotating ^{238}U target wheel. During the experiment, we used two different sets of ^{238}U targets. The beam passed through a 20.2 μm Be vacuum window, 6 mm of He/O_2 gas mixture (He:O_2 = 9/1), a 12.6 μm Be target backing before entering the ^{238}U target (3 segments of 1.8 mg/cm^2, 1.5 mg/cm^2 and 1.6 mg/cm^2). The second target set had a Be backing of 8.2 μm and 3 segments containing 1.0 mg/cm^2 of ^{238}U each. We assumed that only Hs nuclei with a minimum residual range after exiting the target of 10 mm in gas could be transported to the detection system, corresponding to an active layer of the target of 1.0 mg/cm^2. The energy of the ^{36}S ions was in the range of 175.0 MeV to 181.2 MeV within the first set of target and in the range of 190.4 MeV to 196.1 MeV in the second set [6], corresponding to excitation energies, E*, of 38±3 MeV and 51±3 MeV [7], near the predicted maxima of the 4n and 5n evaporation channel, respectively. Starting at E*= 38 MeV we irradiated the first target set with a beam dose of $5.74 \cdot 10^{17}$ ions. The second target set was irradiated with a beam dose of $1.03 \cdot 10^{18}$ ions at E*= 51 MeV.

We searched for correlated decay chains, which were defined as an α-decay ($8.0 \leq E_\alpha \leq 9.5$ MeV) followed within 300 s in the same or a neighbouring detector pair by an α-decay in the same energy range or by a SF-like event with at least one fragment above a threshold of 15 MeV. The data analysis revealed one chain at the higher E* of 51 MeV. A 9.02±0.05 MeV α-particle was observed in bottom detector #24 followed after 23 ms by one 41 MeV fission fragment measured in top detector #24. We attributed this decay chain to the decay of ^{270}Hs produced in the 4n evaporation channel [2].

Because of background from α-decay and SF of heavy nuclides from (multi) nucleon transfer pseudo correlated chains can be found with nonzero probability. We have calculated the probability to observe random decay chains of the types α·α·α·α, α·α·SF, and α·SF satisfying the criteria specified above. 351 and 771 α-particles have been registered in the first and the second run, respectively. They originated mainly from α-decays of ^{212}Po. Also, 6 SF-like events were registered in the first run and 13 events in the second run. None of these were coincident with each other. The results are shown in Table 1.

Table 1: Random rates for different decay chains

decay chain	E* = 38 MeV	E* = 51 MeV
α·α·α·α	$7.08 \cdot 10^{-4}$	$3.12 \cdot 10^{-3}$
α·α·SF	$4.61 \cdot 10^{-3}$	$1.67 \cdot 10^{-3}$
α·SF	$1.6 \cdot 10^{-2}$	$4.6 \cdot 10^{-2}$

At E*= 38 MeV the cross section limit for both channels is 2.9 pb. The cross section for the 4n channel at E*= 51 MeV based on the one event is $8_{-0.7}^{+2.6}$ pb and the cross section limit for the 5n channel is 1.5 pb. Errors and limits correspond to 68% confidence level. The measured cross section and limits are lower than those of the reaction ^{248}Cm(^{26}Mg,xn)$^{274\text{-}x}$Hs [3], in contrast to calculations from [4]. We plan to continue these measurements in the near future.

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First Transactinide Chemistry Behind TASCA*

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The final phase of the TASCA commissioning program [1] included a series of experiments with ^{260}Rf, ^{261a,b}Rf and ^{262}Rf produced in the ^{244}Pu(^{22}Ne,xn) reaction. One of the highlights was a proof-of-principle transactinide chemistry experiment with 78-s ^{261a}Rf. The Automated Rapid Chemistry Apparatus (ARCA) [2] was used to study the formation of fluoride complexes of Rf in diluted HF-solution by anion-exchange chromatography.

TASCA was operated in the High Transmission Mode (HTM) [3] at a pressure of 0.4 mbar He. After passing a 14 cm x 4 cm large Mylar window of 1.2 µm thickness, evaporation residues were thermalized in He at 1200 mbar in a newly designed recoil transfer chamber (RTC) of 1.7 cm depth. The RTC had two funnel shaped inlets at the right and left hand side for the He/KCl jet and a central outlet in the cover plate. The Rf was transported to ARCA by a He/KCl jet through a 10 m long PE capillary of 2 mm i.d. at a gas flow rate of 2.9 L/min. To monitor the gas-jet yield, a ^{227}Ac emanation source was connected to the RTC. Ar with a flow rate of 20 mL/min was passed through the source and transported ^{219}Rn into the RTC. The yield of its decay product ^{211}Bi was compared to the respective yield in ROMA [4]. The ^{211}Bi yield in ARCA including collection, dissolution in $7x10^{-4}$ M HF solution, and evaporation on a Ta disc was 50% of that in ROMA.

For the anion-exchange chromatography in ARCA, the column magazines were filled with the resin MCI GEL CA08Y from Mitsubishi Chemical Corporation, particle size 22±5 µm, which was transferred into the hydroxide form as described in [5]. In each chromatography experiment, two Rf fractions were collected. The first one was $7x10^{-4}$ M HF, which was also used for column loading, and the second one was 5 M HNO_3. The latter was used to strip the remainder of the Rf from the column. After around 18 h experiment, the concentration of the first solution was changed to $1x10^{-3}$ M HF and the experiments were continued for another 25 h.

The KCl clusters were collected in ARCA for 90 s. Within this time, the column for the next experiment was preconditioned for 65 s with the HF solution. After the collection, the products were dissolved in 200 µl of $7x10^{-4}$ M HF solution and were subsequently fed onto the anion-exchange column at a flow rate of 1.0 mL/min. The effluent of the column was collected on a Ta disk as fraction 1. The fraction of the products adsorbed on the resin was eluted with 250 µL of 5 M HNO_3 and collected on a second Ta disk. Both fractions were evaporated to dryness by infrared light and a hot helium stream. The two Ta discs were then subjected to α-spectroscopy. Counting of the first fraction started 60 s after the end of the collection interval, counting of the second fraction started 65 s after the end of the collection.

In total, seven α-events were detected which we attribute to 78-s ^{261a}Rf based on the measured α-energy and lifetime. All of them were observed in the HNO_3 fraction. Two of these events were detected during the experiments with $7x10^{-4}$ M HF, the other five events were detected while using $1x10^{-3}$ M HF. As no events were observed in the HF fractions, it is only possible to give a lower limit for the %ads value. As in Poisson statistics zero observed events are compatible with three events at 95% confidence level, 3 events were assumed for the first fraction and the remainder in the second fraction resulting in %ads ≥ 62.5 % in $7x10^{-4}$ M HF and %ads ≥ 72.5 % in $1x10^{-3}$ M HF. The sum of the α events is shown in Figure 1 indicating that the α spectra were very clean, also thanks to preseparation in TASCA.

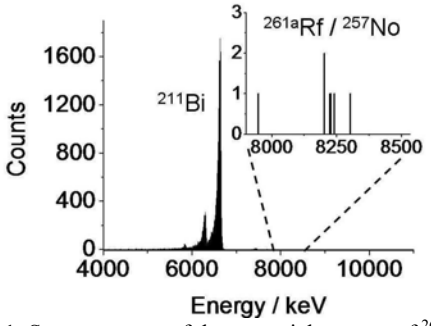

Figure 1: Sum spectrum of the α-particle events of ^{261a}Rf.

From a comparison with the number of ^{261a}Rf events in ROMA, we have to conclude that the chemical yield in ARCA was low, on the order of 30% only. This is in line with earlier observations indicating some sorption of transactinides from HF solutions on the Kel-F slider in ARCA.

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TASCA Commissioning Completed*

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The **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus (**TASCA**) project [1], which is focusing on the separation and investigation of neutron-rich transactinide nuclides produced in actinide-target based reactions, has successfully finished its commissioning; see [2] for an interim report. TASCA is ready for the envisioned research program which includes both chemical investigations of transactinide or superheavy elements (SHE) after preseparation with the gas-filled separator and nuclear structure and nuclear reaction studies.

The central device of TASCA is a gas-filled separator in a DQQ configuration operated either in the "High Transmission Mode" (HTM, DQ_hQ_v) or in the "Small Image Mode" (SIM, DQ_vQ_h) [1-5]. In the HTM, the unsurpassed transmission of TASCA - at a relatively low dispersion - is exploited. In contrast, the SIM provides unique possibilities due to its small spot size in the focal plane (< 3 cm diam.) at a still relatively high transmission; see Table 1.

Table 1: Important parameters of TASCA, calculated for the reaction ^{48}Ca(^{244}Pu,3n)^{289}114, in comparison with other gas-filled separators operated in SHE research.

Separator	Configuration	Transmission %	Dispersion mm/%	Bρ (max) Tm
DGFRS	DQ_hQ_v	35	7.5	3.1
GARIS	DQ_hQ_vD	40	9.7	2.16
BGS	Q_vD_hD	49-59	20	2.5
TASCA	DQ_hQ_v	60	9	2.3
TASCA	DQ_vQ_h	35	1	2.3

Table 2 provides a compilation of all nuclear reactions and reaction products applied and detected in the course of the commissioning program together with the mode TASCA was operated in and the fill gas. Also listed are experiments to test and optimize the recoil transfer chambers (RTC) [6], the gas-jet transport of preseparated products into our Rotating wheel On-line Multidetector Analyzer (ROMA), and its performance, and the coupling and use of aqueous chemistry set-ups behind TASCA.

Table 2: Nuclear reactions and their products used to commission TASCA; H=HTM, S=SIM, TSp=TASISpec, R=ROMA, C=chemistry, catch=catcher foils.

Product	xn	Beam	Target	Mode	Gas	RTC +R/C
^{30}Si	--	^{30}Si	--	H,S	Vac	
^{173,175}Os	7n	^{40}Ar	^{nat}Ce	H	He	C
$^{180-182}$Hg	2-4n	^{40}Ar	^{144}Sm	H,S	He	C
^{188}Pb	4n	^{48}Ca	^{144}Sm	H,S	He	
^{188}Pb	4n	^{40}Ar	^{152}Gd	H,S	He	
$^{194-196}$Pb	4-5n	^{40}Ar	^{nat}Gd	H,S	He	R
$^{198-199}$Bi	4-5n	^{22}Ne	^{181}Ta	H,S	He	catch
$^{195-196}$Po	4-5n	^{48}Ca	^{152}Gd,	H	He	R
^{200}At	3n	^{64}Ni	^{nat}La	TSp	He	
^{200}Fr	5n	^{64}Ni	^{141}Pr	TSp	He	
$^{205-206}$Fr	5-6n	^{30}Si	^{181}Ta	H	He	
$^{208-211}$Ra	3-4n	^{54}Cr	^{nat}Gd	H,S	He	
$^{208-211}$Ra	3-6n	^{64}Ni	^{150}Nd	TSp	He	
^{210}Ac	5n	^{40}Ar	^{nat}Lu	H,S	He,N_2	
^{215}Ac	4n	^{22}Ne	^{197}Au	H,S	He,H_2	
$^{218-x}$Th	xn	^{64}Ni	^{154}Sm	TSp	He	
$^{224-x}$U	xn	^{64}Ni	^{nat}Gd	TSp	He,H_2	
^{245}Fm	3n	^{40}Ar	^{208}Pb	H,S	He	R
^{252}No	2n	^{48}Ca	^{206}Pb	H,S, TSp	He	R
^{253}No	2n	^{48}Ca	^{207}Pb	H, TSp	He	
^{254}No	2n	^{48}Ca	^{208}Pb	H,S	He,H_2	
^{255}No	5n	^{22}Ne	^{238}U	H,S	He,H_2	R
^{256}No	4n	^{22}Ne	^{238}U	H	He	
^{260}Rf	6n	^{22}Ne	^{244}Pu	H	He,H_2	
^{261a,261b}Rf	5n	^{22}Ne	^{244}Pu	H	He	R,C
^{262}Rf	4n	^{22}Ne	^{244}Pu	H	He,H_2	

Extensive studies have been performed in the HTM and SIM to obtain optimized parameter sets for (i) the target thickness and stability, (ii) the gas pressure and the gas filling (He, H_2, and mixtures), (iii) the dipole setting (Bρ) and quadrupole focusing, (iv) the RTCs (window material and thickness, support structures, and size and shape of the chamber), (iv) gas-jet transport of pre-separated products, and (vi) the coupling and performance of devices

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like ROMA and the Automated Rapid Chemistry Apparatus (ARCA). Results of many of these parameter studies were compared with TASCA model calculations [7] and very good agreement was achieved. This agreement is of special importance as it allows for the selection of proper settings for magnetic rigidities (Bρ) in the dipole magnet and the quadrupole magnets for all nuclear reactions and for all gases and gas mixtures tested at various pressures. It is especially rewarding to see that not only Bρ values were properly chosen to centre product distributions on focal plane detectors (FPD) but also that the measured spatial distributions and, more importantly, the efficiencies were in very good agreement with model calculations. These results confidently demonstrate that we are able to perform trustworthy SHE experiments with TASCA. In the following, we mention a few concluding experiments, some of the highlights and new developments; see [2] for additional information on the parameter studies.

The first efficiency measurements with catcher foils behind the target and in the focal plane showed very good agreement with model calculations for the fairly asymmetric reaction ^{22}Ne(^{181}Ta,xn)^{198m,199}Bi [2]. To confirm this agreement in a more symmetric reaction, leading to a significantly heavier reaction product, and to obtain a standard reaction to test and check the TASCA performance, detailed studies were performed with well known reactions of ^{48}Ca with ≈ 0.5 mg/cm^2 thick targets of ^{206,207,208}Pb leading to ^{252,253,254}No. Assuming cross sections of 0.5 µb, 1.3 µb, and 2 µb [8] for the production of ^{252}No, ^{253}No, and ^{254}No, efficiencies of 54%, 56%, and 50%, respectively, were obtained for the HTM, using a He filling of 0.8 mbar, and a (80x36) mm^2 16-strip FPD. Taking into account uncertainties in cross sections and systematic errors of target thicknesses and beam current measurements, we observe an excellent agreement with model calculations [7] predicting 54%. Equally good is the agreement in the SIM, at a He pressure of 0.8 mbar, where a 30% efficiency was measured for the reaction ^{48}Ca(^{208}Pb,2n)^{254}No.

A new (140x40) mm^2 large, highly efficient FPD, consisting of double-sided silicon strip detectors (DSSSD) in the focal plane and SSSDs for the backward box detectors will further increase the TASCA efficiency; see [9] for details of the new detector.

As one of the crucial tests and one of the highlights finalizing the TASCA commissioning program, we studied the isotopes ^{260}Rf, ^{261a,261b}Rf, and ^{262}Rf synthesized in the very asymmetric reaction ^{22}Ne + ^{244}Pu; see [10] for details of the nuclear reactions, for TASCA parameters, and for the interesting nuclear decay results. In essence, the performance of TASCA was as anticipated; everything worked well, including the ^{244}Pu target wheel. Efficiencies and magnetic settings (Bρ=1.99 Tm at 0.4 mbar He, HTM) were as expected. As observed in previous experiments [2], it was again possible to reduce the background in the FPD by using a mixture of He and H_2. This part of the commissioning program showed clearly that TASCA can be applied efficiently for nuclear decay and nuclear reaction studies of neutron-rich nuclides of SHE synthesized in very asymmetric hot-fusion reactions. Rf isotopes were not only measured in the FPD but were also collected in an RTC and were transported either to ROMA for nuclear decay measurements [10] or to ARCA for chemical investigations [11].

An additional highlight of the experiment was the first transactinide chemistry behind TASCA designed as a proof-of-principle experiment. It was performed in ARCA with pre-separated 78-s ^{261a}Rf; details of the nuclear reaction and the Rf separation in TASCA are described in [10] while all chemical aspects are discussed in [11]. This successful experiment, which studied the formation of Rf-fluoride complexes and their adsorption behaviour on an anion-exchange resin, demonstrated that aqueous-phase transactinide chemistry behind TASCA can now be performed.

The new set-up termed *TA*sca *S*mall *I*mage mode *Spec*troscopy (TASISpec) [12] exploits advantages of the SIM, i.e. the fact that neutron-rich nuclides of SHE, produced in hot-fusion reactions, can be focused with high efficiency into an area of < 7 cm^2. This provides the unique possibility to build a compact Si-detector box for α-particle, electron, and fission-fragment measurements, and to pack composite Ge-detectors in very close geometry, resulting in an unprecedented, highly efficient set-up for multi-coincidence measurements with γ-rays and X-rays; see [12] for details. A prototype set-up has been commissioned successfully and first data have been collected for nuclides as heavy as ^{252,253}No.

In conclusion, the performance of TASCA as a separator is well understood and is perfectly under control. TASCA as a whole is presently the most versatile and highest efficient instrument in SHE research worldwide. It has entered the region of transactinides or superheavy elements, and is ready to explore the physics and chemistry of the "terra incognita" it was designed and built for.

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Measurements of $^{260\text{-}262}$Rf produced in ^{22}Ne + ^{244}Pu fusion reaction at TASCA*

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As a final experiment in the commissioning phase of TASCA the transactinides (Z ≥ 104) were reached. Production and decay of ^{260}Rf, ^{261a,b}Rf and ^{262}Rf [1,2,3], produced in the asymmetric nuclear fusion reaction ^{244}Pu(^{22}Ne,xn) was studied. Separated reaction products were guided to a Focal Plane Detector (FPD) or into a Recoil Transfer Chamber (RTC), where they were available for transport to either the Rotating wheel On-line Multidetector Analyzer (ROMA) or to the Automated Rapid Chemistry Apparatus (ARCA) for chemical experiments [4].

TASCA was operated in the High Transmission Mode (HTM) [5]. The ^{22}Ne ion beam (average intensity: 0.8 μA_{part}) impinged on a rotating target wheel with 0.4 mg/cm^2 $^{244}PuO_2$ targets on 2.2 µm Ti backings. Three beam energies in the center of the target, $E_{c.o.t.}$, of 109 MeV, 116 MeV and 125 MeV, were used for the production of ^{262}Rf, ^{261}Rf and ^{260}Rf, respectively. The transmission of Rf has been optimized in He filling gas. The optimal pressure was 0.4 mbar. The magnetic rigidity, Bρ, was determined to be 1.99 T·m. To increase suppression of unwanted products, a He/H_2 (2:1) filling gas at a pressure of 1.5 mbar was used in experiments with the FPDs. Evaporation residues were implanted into a (80 x 36) mm^2 16-strip Position-Sensitive silicon Detector (PSD) or a (58 x 58) mm^2 Double-Sided Silicon Strip Detector (DSSSD). In other experiments, ^{261a,b}Rf passed a 1.2 µm thick (140 x 40) mm^2 Mylar window and was thermalized in 1.2 bar He in the RTC (depth: 17 mm). Rf atoms were then transported to ROMA by an He/KCl jet (gas flow rate: 3.45 L/min) through a 4 m long polyethylene capillary (inner diameter: 2 mm).

The measurement of ^{260}Rf, produced in the 6n evaporation channel at $E_{c.o.t.}$= 125 MeV yielded 15 time (Δt ≤ 200 ms) correlated EVR-SF events in the PSD. The correlation time analysis yielded a half-life of $21^{+7.3}_{-4.3}$ ms (errors are within the 68% confidence interval). A search for ^{262}Rf decays at $E_{c.o.t.}$ = 109 MeV yielded 7 position and time correlated EVR-SF events observed in the DSSSD, with EVR energies of 0.8 to 3.3 MeV and SF fragment energies of > 100 MeV. The measured $T_{1/2}$ for ^{262}Rf is 210^{+128}_{-58} ms (Fig. 1a), in contradiction with values from [1,2]. In addition, 9 short EVR-SF correlations were registered with Δt ≤ 1.5 ms and EVR energies of 7.5 ± 5.0 MeV. They were attributed to the decay of ^{244mf}Am ($T_{1/2}$ = 0.9 ms). Because of a relatively high counting rate of EVR-like events in the DSSSD a random event analysis was performed for EVR-SF correlations within a Δt of 1 s. The random event number, n_b, was calculated individually for each observed event. It varies between 0.035 and 0.11 and depends on the event position in the DSSSD.

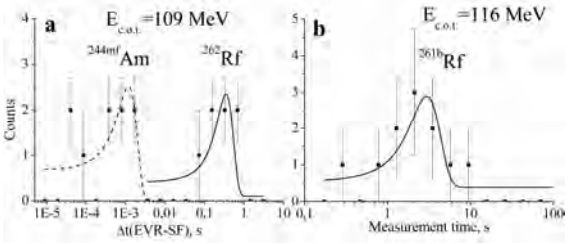

Figure 1: Time distributions of a) EVR-SF correlations from the DSSSD and b) SF decays from ROMA.

^{261}Rf was produced in the 5n channel at $E_{c.o.t}$ = 116 MeV and was detected in ROMA. Stepping times of 35 s (for ^{261a}Rf) and 2.5 s (for ^{261b}Rf) were used. 149 single α-particles (E_α = 7.8 – 8.5 MeV) from ^{261a}Rf and ^{257}No were registered; among these 28 α-α correlations. Also, 11 SF-events were registered and are attributed to ^{261b}Rf based on the measured $T_{1/2}$ of $2.2^{+0.9}_{-0.5}$ s. The SF activity assigned in [2] to ^{262}Rf likely originated from the then unknown ^{261b}Rf.

From our results and a cross section of 4.4 nb [6], a transmission of Rf through TASCA to a 140 x 40 mm^2 large area in the focal plane of 10% follows. For ^{261b}Rf, a cross section of $1.8^{+0.8}_{-0.4}$ nb was calculated, respecting decay during transport. With an estimated transmission of 6% to the area of the FPDs, preliminary cross sections for ^{260}Rf and ^{262}Rf of ≈ 1.2 nb and ≈ 250 pb, respectively, follow. ^{261b}Rf was observed for the first time as an EVR. The production ratio of ^{261a}Rf to ^{261b}Rf is about 2.5:1. The data analysis is still in progress.

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A new TASCA focal plane detector*

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The new, highly efficient, gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) has been recently put into operation at GSI with the aim to study chemical and physical properties of superheavy elements with Z≥104. Based on the results of magnetic field model calculations of the dipole and the quadrupoles, two modes of operation of TASCA, the "High Transmission Mode" and the "Small Image Mode" have been realized [1]. Two types of focal plane detector setups (FPD) were used during the TASCA commissioning phase: a (80 x 36) mm^2 16-strip position-sensitive silicon detector (PSD) and a (58 x 58) mm^2 double-sided silicon strip detector (DSSSD) as a prototype for a new TASCA focal plane detector. The test experiments showed that the image size in the HTM is larger than the detector size of both detector types, as it was expected according to ion transport calculations and Monte-Carlo simulations [1,2]. The nominal vertical position resolution of the PSD is ± 0.2 mm. However, a search for position correlated decay chain members usually occurs within ± 1 mm limits, corresponding to a pixel size area of (5 x 2) mm^2. The relatively large pixel size of the PSD negatively affects search limits for rare decay chains from long-lived isotopes of superheavy elements. The TASCA FPD working group has decided to build a detector setup based on a DSSSD as a stop detector and a backward array consisting of single-sided silicon strip detectors (SSSD). A *veto* detector for light fast ions that penetrate the DSSSD will be mounted behind the stop detector. The new setup will feature a (144 x 489 mm^2 large detector, which will accept > 90% of all evaporation residues reaching the focal plane. The geometrical detection efficiency for α-particles emitted from implanted nuclei will be > 70%. As compromise between a pixel size as small as possible and the number of spectrometric electronic channels as small as possible, a pitch width of 1 mm on the front and back side of the DSSSD has been chosen. To reduce the necessary number of ADCs the number of strips on each side of the DSSSD and on the SSSD should be a multiple of 8 when using 32-channel preamplifiers and 8 or 16-channel amplifiers with integrated multiplexers. A DSSSD structure with an active area of (72 x 48) mm^2 has 72 strips on the front side and 48 strips on the back side. The strip width and the interstrip distance on the both sides of the DSSSD are 900 μm and 100 μm, respectively. Two adjacent DSSSDs form the stop detector with an active area of (144 x 48) mm^2. A SSSD structure with an area of (72 x 48) mm^2 has 8 strips, which are 72 mm long. The strip width and the interstrip distance of the DSSSD are 5.65 mm and 100 μm, respectively. 8 SSSD detectors form the backward array with a depth of 72 mm. Two similar SSSD detectors are used as *veto* detector. Silicon wafer thicknesses of 300 μm and 500 μm have been chosen for the DSSSDs and SSSDs, respectively. These thicknesses will allow for the detection of conversion electrons in the backward array. A schematic design of the new TASCA detector array and first results from source measurements are shown in Fig. 1.

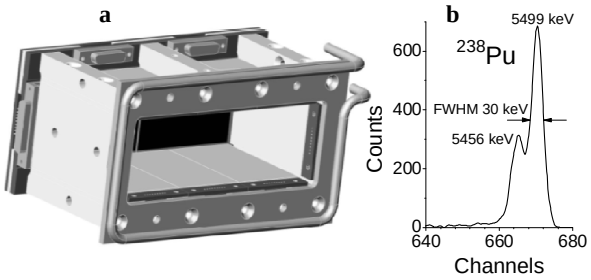

Figure 1: A drawing of the new TASCA FPD array (a) and an alpha particle spectrum from a DSSSD strip (b).

In total, the new TASCA detector array has to process the signals of 320 strips. The necessary spectrometric and data readout electronics, which are compatible with TASISpec [3], as well as the associated software, should be ready for experiments in spring 2009.

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TASISpec - A new twist on spectroscopy of superheavy elements.

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A new nuclear spectroscopy set-up called TASISpec (*TA*sca *S*mall *I*mage mode *Spec*troscopy) has been designed and commissioned. It exploits TASCA's specific small image focal mode, i.e. the fact that superheavy elements (SHE) produced in fusion-evaporation reactions can be focused into an area of less than 3 cm in diameter [1]. This provides the world-unique possibility to pack composite Ge-detectors in very close geometry, resulting in an unprecedented, highly efficient detection of γ-rays and X-rays in coincidence with implanted SHE.

The test set-up as used in 2008 is illustrated in Fig. 1. It comprised a focal plane made from a 58 mm $\times$ 58 mm large double sided silicon strip detector (DSSSD) complemented by four single sided silicon strip detectors (SSSD) mounted in the backward hemisphere and two Ge detectors: one cluster (7 crystals) downstream and one large VEGA clover (4 crystals) on one side. The final set-up foresees three additional clover detectors, while their implementation requires a dedicated holding structure to be built in 2009.

Conversion electron, γ-ray, and α-particle sources as well as evaporation residues from the reactions ^{206}Pb(^{48}Ca, $2n$)^{252}No, ^{207}Pb(^{48}Ca, $2n$)^{253}No, ^{244}Pu(^{48}Ca, $4n$)^{288}114, ^{150}Nd(^{64}Ni, xn)$^{214-x}$Ra, ^{154}Sm(^{64}Ni, yn)$^{218-y}$Th, and ^{nat}Gd(^{64}Ni, zn)$^{224-z}$U have been used to characterise the performance of TASISpec. Detection efficiencies of some 80% were measured for emitted α particles and extrapolated to more than 40% (absolute scale) for γ rays at an energy around 250 keV in the final set-up. In addition, energy thresholds of particle- and γ-ray detectors, dead times, and data rates of a first combined VME (up to 224 channels for Si-strip detector processing) and XIA-DGF (11 channels sampling the Ge-detector signals) data acquisition system were successfully tested as well as TASCA-SIM transmissions determined at different magnet settings.

In 2009 it is planned to establish the extraordinary $\gamma\gamma$-, γ-CE-, CE-CE, or even $\gamma\gamma$-CE-, multi-coincidence capabilities of the both segmented and compact TASISpec set-up by settling the decay scheme of K-isomeric states in ^{253}No [2, 3, 4, 5]. Despite of the underlying amount of data many question marks still remain regarding the structure of ^{253}No.

Following upon this ideal starting point to show the proof-of-principle of TASISpec, K isomers expected in neutron-rich SHE will be stepwise approached. Such a programme employs also the unique facets of both high UNILAC beam intensities, use of radioactive actinide targets, and high transmission of TASCA for rather asymmetric reactions. Last but not least, the unprecedented γ-efficiency of TASISpec may allow to identify SHE by means of characteristic X-rays.

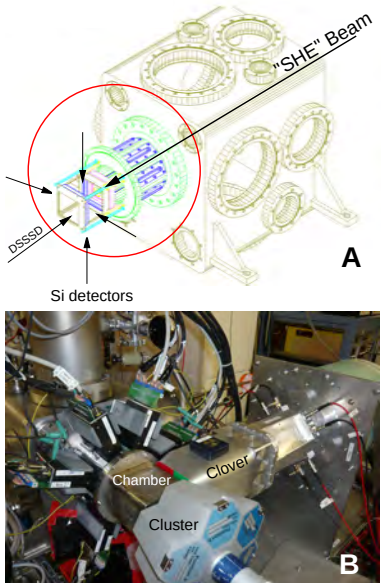

Figure 1: The TASISpec set-up installed at the focal plane of TASCA. Panel (A) illustrates a drawing of the focal-plane area of the TASCA separator. The TASISpec structure is encircled. Five silicon strip detectors are placed in a cube-like shape. Panel (B) shows a photograph of the commissioning set-up. The relative positions of the Ge-VEGA-clover and Ge-cluster detector is here visible.

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Calculation of Adsorption Energies of Elements 112 and 114, and their Homologues Mercury and Lead on Gold (111) Surface

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In the last decade the gas-phase thermography becomes the method of choice for studying the chemical behavior of super-heavy elements in nuclear-chemistry. In such experiments, however, the whole information about the adsorption process of a chemical element is reduced to a single number – the adsorption temperature. The adsorption on the other hand is a very complex process, which requires an adequate theoretical description for interpreting the experimental data.

Within our theoretical approach, which is based on fully-relativistic 4*c*-DFT [1], the substrate surface is represented by a cluster of atoms at fixed positions. The adsorption behaviour, i.e. binding energies and structures strongly depend on the size of the chosen cluster. Therefore a cluster size convergence studies are mandatory in order to be able to obtain accurate results.

In this report we present our calculations of the adsorption energies of elements 112 and 114 and their homologues Hg and Pb on a gold (111) surface for different cluster sizes. For all four possible adsorption positions – top, bridge, hollow1, and hollow2 – we first performed calculations for the smallest possible cluster, which for the on top site might even be a single atom only, for example. In the next step we added all atoms from the next coordination shell to our cluster and repeated the calculations. We continued this procedure until convergency was achieved in both the adsorption energy and adsorption distances. The results of this study are summarized in Figure 1, where the potential energy curves for elements 112 and 114, and mercury are presented. (Pb is not shown in the picture, because the binding energies are too large). The convergence of both the bond distance and the adsorption energy as function of cluster size was achieved with the largest clusters containing 95 (top), 94 (bridge), 120 (hollow1), and 107 (hollow2) gold atoms.

The results of the calculations show that the binding energies of each of the considered elements for the bridge and hollow2 positions are very similar. However, element 114 and Hg have a slight preference to adsorb in the bridge position, while element 112 and Pb - in the hollow2 one. Except for Pb, all the other three elements are weakly bound to the gold surface which is due to the strong relativistic effects in the outermost shells of these elements and a closed shell effect in Hg and element 112.

An interesting and important feature of the present calculations is that the binding energies change with both the adsorption position and cluster size in such a way that the adsorption strength order $E_b(E112) < E_b(Hg) < E_b(E114) \ll E_b(Pb)$ remains the same. This confirms our former qualitative predictions [2], which were based on calculations for dimers only. A detailed discussion of the presented results can be found in [3].

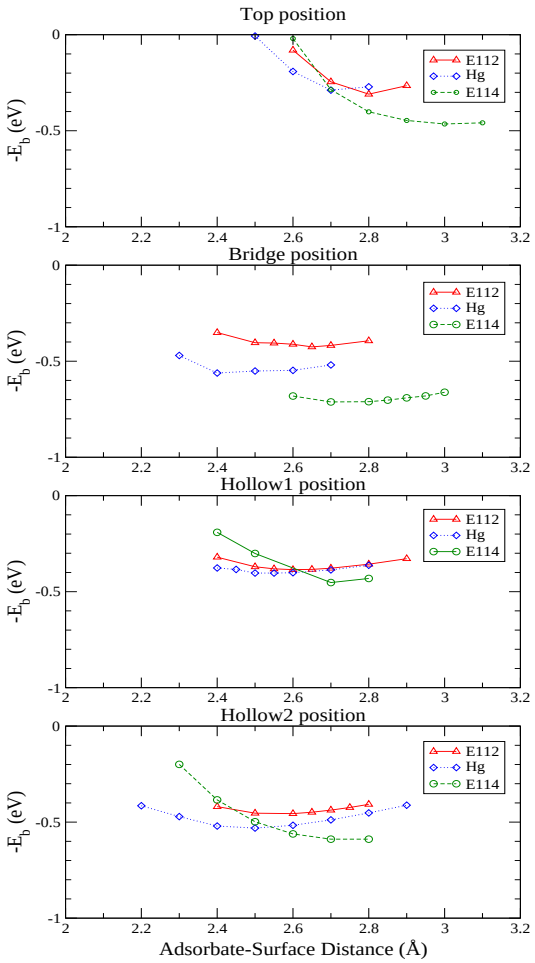

Figure 1: Adsorption energies of elements 112, 114, and mercury as function of the adsorbate-surface distance.

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Prediction of Adsorption of Element 113 on Inert Surfaces from *ab initio* Dirac-Coulomb Atomic Calculations

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Element 113 has an isotope (A=284) with $t_{1/2}=0.48^{+0.58}_{-0.17}$ s, which makes it suitable for chemical studies. The element is expected to be volatile. Its adsorption behaviour is to be investigated by gas-phase chromatography experiments using silicon detectors of the chromatography column covered with gold layers. Feasibility experiments are under way, studying the adsorption behaviour of the nearest homolog, Tl [1]. Prediction of the adsorption enthalpy, ΔH_{ads}, and temperature, T_{ads}, of element 113 on gold surface is very important for the planned experiments. Information about adsorption on inert surfaces such as Teflon and polyethylene (PE) is also valuable, as these materials are used as transport capillaries from the target chamber to the detection system in the experimental setup. In this work, we predict the adsorption behaviour of element 113 and its homolog Tl on Teflon and PE on the basis of very accurate results of *ab initio* calculations of their atomic properties.

The electronic structure calculations were performed using the DIRAC package [2]. In the Dirac-Coulomb (DC) *ab initio* method, the many-electron relativistic Dirac-Coulomb Hamiltonian

$$H_{DC} = \sum_i h_D + \sum_{i<j} 1/r_{ij} \qquad (1)$$

is employed, where

$$h_D = c\vec{\alpha}\cdot\vec{p} + \beta c^2 + V_{nuc}\,. \qquad (2)$$

The atomic orbitals (AO) are four-component spinors

$$\phi_{nk} = \begin{pmatrix} P_{nk}(r) \\ Q_{nk}(r) \end{pmatrix}, \qquad (3)$$

where $P_{nk}(r)$ and $Q_{nk}(r)$ are the large and small component, respectively. The Faegri uncontracted 26s24p18d13f5g2h basis set was used for both elements [3]. Electron correlation was taken into account at various levels of theory including the highest, the Fock-Space Coupled Cluster Single Double excitations, FS CCSD, for which the current results are presented.

The calculations of polarizability (α) were performed by the finite field method. The DC FS CCSD results for α are given in Table 1 along with the ionization potentials (IP) of Tl and element 113 calculated at best using the Dirac-Coulomb-Breit (DCB) FSCC method [4]. The van der Walls radii (R_{vdW}) were determined from a linear correlation with $R_{max}(np_{1/2})$-AOs of the group 13 elements. ΔH_{ads} were calculated using a model of physisorption given by eq. (8) of Ref. [5].

Table 1. Atomic properties of Tl and element 113: ionization potentials, IP (in eV), polarizabilities, α (in a.u.), van der Walls radii, R_{vdW} (in Å) and adsorption enthalpies, ΔH_{ads} on Teflon (T) and PE (in kJ/mol).

Property	Tl	Meth.	Ref.	113	Meth.	Ref.
IP	6.108	DCB	[4]	7.306	DCB	[4]
	6.110	exp.	[6]	-	-	
α	51.3	DC	this	29.85	DC	this
	51(7)	exp.	[6]	-	-	
R_{vdW}	1.90	corr.	this	1.84	corr.	this
$-\Delta H_{ads}$(PE)	22.22	calc.	this	15.83	calc.	this
$-\Delta H_{ads}$(T)	19.65	calc.	this	14.00	calc.	this

The obtained ΔH_{ads} on PE and Teflon for all group 13 elements are shown in Fig. 1, revealing a reversal of the trend beyond In due to the relativistic contraction of the $np_{1/2}$ AO. The very low $-\Delta H_{ads}$(113) on inert materials guarantees its transport to the chemistry set up.

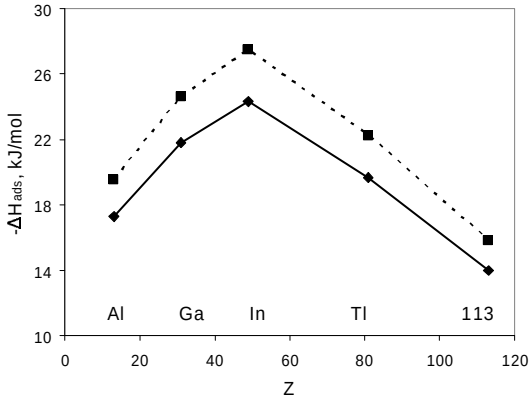

Fig. 1. Calculated adsorption enthalpies of group 13 elements on PE (dashed line) and Teflon (solid line).

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Relativistic *ab initio* Study of HgAu, Homologue of the Superheavy 112Au

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Predictions of the interaction of the superheavy elements with various surfaces are essential for their identification and separation. Recently, we have predicted the adsorption properties of the superheavy element 112 on inert surfaces [1]. The calculations were performed using the fully relativistic Dirac-Coulomb Hamiltonian combined with the coupled cluster method. The accuracy of the results was verified by performing the same calculations on mercury, which is a lighter homologue of element 112, and for which experimental data exist. For Hg, the results obtained were in excellent agreement with the experimental values, and we expect the same accuracy for its superheavy homologue. Similar investigations were performed on element 114 and Pb.

In this work we investigate the adsorption of superheavy element 112 and Hg on gold through the fully relativistic *ab initio* electronic structure calculations for the dimers 112Au and HgAu. The calculations have been completed for HgAu. Those for 112Au are under way.

The electronic structure calculations are performed using the DIRAC package [2]. In order to take relativity into account, we employ the 4-component Dirac–Coulomb Hamiltonian,

$$H_{DC} = \sum_i h_D + \sum_{i<j} 1/r_{ij}, \quad (1)$$

where

$$h_D = c\vec{\alpha}\cdot\vec{p} + \beta c^2 + V_{nuc}. \quad (2)$$

V_{nuc} is the nuclear attraction operator, and α and β are the four-dimensional Dirac matrices. In order to obtain computational efficiency, the interatomic SS-integral contributions were modeled by classical repulsion of small component atomic charges [3]. This is one of the most economical and accurate approximation to the full Dirac-Coulomb Hamiltonian. Electron correlation is taken into account using the Fock space coupled cluster (FS CC) method, which is considered to be one of the most powerful tools in quantum chemistry.

The basis set of Saue *et al* [4], consisting of 23*s*, 18*p*, 14*d*, and 8*f* orbitals, was used for the gold atom. For the mercury atom the basis set of Visscher [5] was used, with 20*s*, 18*p*, 12*d*, and 10*f* orbitals.

HgAu is an open shell system; hence we start our calculation with the closed shell reference state, $HgAu^-$. After solving the Dirac-Fock equations and correlating the closed shell reference state, one electron is removed, to give the neutral system. After recorrelation, an additional electron is removed, and the positive ion $HgAu^+$ is reached. Thus, in a single calculation we obtain the energies of HgAu, $HgAu^+$, and $HgAu^-$. The calculations were repeated for different values of the interatomic distance, to obtain the potential curves of the systems under study. The spectroscopic properties of interest were obtained from the potential energy curves, using the LEVEL program package [6].

To our knowledge, no experimental spectroscopic data exist for the HgAu molecule. However, this system has been extensively studied theoretically [7-11], mostly using relativistic DFT methods. A relativistic coupled cluster single doubles (triples) (RCCSD(T)) investigation was also performed [10]. A comparison with the other calculations is given in Table 1. One can see very good agreement with the 4*c*-DFT result for HgAu [11].

Table 1. Spectroscopic properties of HgAu, $HgAu^+$, and $HgAu^-$: equilibrium bond lengths R_e, (in Å), dissociation energies, D_e, (in eV), adiabatic ionization potentials, IP_{ad},(in eV), and vibrational frequencies, ω_e, (in cm^{-1})

Property	AuHg	$AuHg^+$	$AuHg^-$	Method, Ref.
R_e	2.657	2.553	2.854	DC FSCC, this
	2.711	2.581	2.967	RCCSD(T) [10]
	2.67	-	-	4*c*-DFT [11]
D_e	0.478	1.712	0.520	DC FSCC, this
	0.389	1.856	0.381	RCCSD(T) [10]
	0.495	-	-	4*c*-DFT [11]
IP_{ad}	7.374	-	1.995	DC FSCC, this
ω_e	116	156	79	DC FSCC, this
	103	139	64	RCCSD(T) [10]

Our results are expected to be more accurate than the RCCST(T) ones due to the larger basis set employed. The same degree of reliability may be expected for the heaviest homolog of HgAu, the 112Au molecule.

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