



# TASCA 18

GSI, Darmstadt, September 25, 2018  
17<sup>th</sup> Workshop on  
Recoil Separator for Superheavy Element Chemistry

supported by the Helmholtz Institute Mainz and the JAEA Reimei Program

**Location: Seminar Room SB3 3.170a**

Last update: 20 Sept. 2018

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All presentations include 20 minutes presentation plus 5 minutes discussion time



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## Recent Results from the FIONA Separator at LBNL

J.M. Gates

*Lawrence Berkeley National Laboratory, Berkeley, USA*

Recently, the Berkeley Gas-filled Separator (BGS) at the Lawrence Berkeley National Laboratory (LBNL) was coupled to a new mass analyzer, FIONA. The goal of BGS+FIONA is to provide a  $M/\square M$  separation of  $\sim 300$  and transport nuclear reaction products to a shielded detector station on the tens of milliseconds timescale. These upgrades will allow for direct  $A$  and  $Z$  identification of ii) new actinide and transactinide isotopes with ambiguous decay signatures such as electron capture or spontaneous fission decay and i) superheavy nuclei such as those produced in the  $^{48}\text{Ca}$  + actinide reactions. Here we will present recent results from the FIONA commissioning and first scientific experiments.

Financial Support was provided by the Office of High Energy and Nuclear Physics, Nuclear Physics Division, and by the Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences and Biosciences of the U.S. Department of Energy, under Contract No. DE-AC02-05CH11231.

## MRTOF Mass measurements at RIBF: Recent measurements of heavy isotopes and future plans for the super-heavy region

M. Rosenbusch for the SHE-mass collaboration of RIKEN-KEK

*RIKEN Nishina Center, Wako-shi, Japan*

Within the SHE-mass collaboration of RIKEN and KEK, first mass measurements of Md isotopes [1] and many other radioactive species like Ac/Ra isotopes [2] have been performed using a multi-reflection time-of-flight spectrograph (MRTOF-MS) [3]. After these successful experiments, the MRTOF-MS has changed the location behind RIKEN's RRC accelerator. Our new aim is, to determine the masses and atomic numbers of  $^{284}\text{Nh}$  and  $^{288}\text{Mc}$  for the first time, which are isotopes in the SHE region disconnected from well-known isotopes by the dominance of spontaneous fission. Furthermore, new MRTOF-MS devices are planned to perform mass measurements of the most exotic species produced at RIKEN. Those devices will be placed in various locations as behind RIKEN's zero-degree spectrometer for accessing exotic nuclides in symbiotic operation with other experiment. In this contribution, an overview of the actual status and future plans for low-energy precision mass measurements will be discussed.

### References

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## Low-lying states in $^{219}\text{Ra}$ and $^{215}\text{Rn}$ : sampling microsecond $\alpha$ -decaying nuclei

A. S  mark-Roth, on behalf of the TASCA E115 Collaboration

*Department of Physics, Lund University, Lund, Sweden*

Short-lived  $\alpha$ -decaying nuclei ‘North-East’ of  $^{208}\text{Pb}$  in the chart of nuclides were studied using the reaction  $^{48}\text{Ca} + ^{243}\text{Am}$  with the decay station TASISpec at TASCA, GSI Darmstadt. Decay energies and times from pile-up events were extracted with a tailor-made pulse shape analysis routine and specific  $\alpha$ -decay chains were identified in a correlation analysis. Decay chains starting with the even-even  $^{220}\text{Ra}$  and its odd-A neighbors,  $^{219}\text{Fr}$ , and  $^{221,219}\text{Ra}$ , with a focus on the  $^{219}\text{Ra} \rightarrow ^{215}\text{Rn}$  decay, were studied by means of  $\alpha$ - $\gamma$  spectroscopy.

As a main result, an  $\alpha$ -decay branch from the excited state at 17 keV in  $^{219}\text{Ra}$  is proposed to resolve a discrepancy between the evaluated level scheme and the experiment. This 17-keV state in  $^{219}\text{Ra}$  was unknown in previous decay spectroscopy experiments. Our results are consistent with Geant4 simulations, and further support the debated ground-state spin of  $^{219}\text{Ra}$ , its first excited state, as well as the low-lying levels in  $^{215}\text{Rn}$ . Contemporary theoretical calculations on deformation, rotational states and the  $\alpha$  decay justify the nuclear structure interpretation. Furthermore, the results show that the measured half-lives differed significantly in the cases of the  $^{216,217}\text{Rn}$ ,  $^{219}\text{Fr}$ ,  $^{215}\text{At}$  and  $^{221}\text{Ra}$   $\alpha$  decays and a revision of the evaluated half-lives – from measurements made before 1970 – is suggested.

## Present status of SHE chemistry at JAEA

T. K. Sato

*Advanced Science Research Center, Japan Atomic Energy Agency, Japan*

It has been pointed out that periodicity of chemical properties is likely to be violated in the superheavy element (SHE) region. The reason for this violation is due to a rearrangement of outermost electrons by strong relativistic effects in some cases. It can be anticipated that atomic properties of SHE would be sensitive on the influence of relativistic effects because electrons in the rearranged outermost electronic orbital play a role in chemical behavior of the atom.

At JAEA, we conducted successful measurements of the first ionization potentials of heavy actinides, fermium (Fm,  $Z = 100$ ), mendelevium (Md,  $Z = 101$ ), nobelium (No,  $Z = 102$ ), and lawrencium (Lr,  $Z = 103$ ) using a surface ionization method [1,2]. As the next step, we have begun to study adsorption behavior of Lr on a metallic surface as an application of the surface ionization method. In addition, on a basis of the system, we have been developing an atomic beam source which can be applicable to directly determine the ground-state electronic configuration of Lr. In parallel, we have just started to develop a novel method to measure the first ionization potentials of superheavy elements heavier than Lr.

In the presentation, the current status of SHE chemistry at JAEA will be presented and a future plan will be briefly discussed.

## References

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## Electron-capture delayed fission in the heaviest nuclei

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Radioactive decays of the heaviest nuclei provide unique information's on the limit for existence of the matter. In this regards, the subsequent decays of the heaviest elements leads to the production of the most stable isotopes of the elements that exist in the nature. A special interest is the so called beta/electron captured (EC)-delayed fission process. EC delayed fission (ECDF) is a two-step process where first mother odd-odd nucleus undergoes EC decay and form an excited daughter even-even ones which directly fission instead of surviving. Despite termination, ECDF has a great impact on the understanding of nature, e.g., isotopic distribution of the astrophysics processes, gives an access to fission from excited states, etc..

Presently, more than 30 cases of ECDF in the isotopes of elements Tl, Bi, At, Fr, Np, Am, Bk, Es and Md are known. ECDF process often quantified by its probability ( $P_{\text{ECDF}}$ ), expressed as a ratio of numbers of initial EC decay and subsequent fission from the excited states of the daughter nucleus, which is still poorly describable by theories. Therefore, the experimental  $P_{\text{ECDF}}$  values have mostly been used in the theory either for extraction of the fission barrier heights or suggestive ECDF half-lives. These approaches lead to interesting results, which help to accumulate the knowledge on ECDF process. Despite these valuable results, still no conclusive picture/view on ECDF that provides a quantitative description of the experimental  $P_{\text{ECDF}}$  values is yet given. Accordingly, theoretical predictions of the yet unknown cases of the ECDF in wide ranges of Z and N do not exist yet.

I will present, a semi-empirical estimate on ECDF probabilities of nuclei with  $Z=79-119$  by inferring the theoretical  $Q_{\text{EC}}$  and  $B_f$ .

## Recent results of theoretical studies of properties of elements 113 through 115

V. Pershina<sup>1</sup>, M. Iliaš<sup>2,3</sup>

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Interesting gas-phase chromatography experiments were conducted on volatility of Cn and Fl as adsorbed on gold and other surfaces [1-4]. The first attempt of chemical characterization of Nh has also been announced using a similar approach [5]. To render assistance to those experiments, relativistic calculations of interaction of those elements and their homologs with gold and other surfaces have been performed using advanced periodic codes [6,7]. Moreover, these theoretical studies were extended to predictions of properties of Nh compounds [8], that can be formed at the experimental conditions, and of their interaction with the surfaces. Calculations for smaller systems of Mc in view of future gas-phase experiments are also presented.

### References

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- [3] A. Yakushev, et al. Inorg. Chem. 53, 1624 (2014).
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## Penning-Trap Mass Spectrometry of the Heaviest Elements with SHIPTRAP

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Ch. E. Düllmann<sup>1,2,6</sup>, M. Eibach<sup>9</sup>, S. Eliseev<sup>5</sup>, P. Filianin<sup>5,7</sup>, S. Götz<sup>6</sup>, Y. Gusev<sup>7</sup>, M. Gutierrez<sup>10</sup>,  
F. P. Hessberger<sup>1,2</sup>, O. Kaleja<sup>5,6</sup>, J. van de Laar<sup>1,6</sup>, M. Laatiaoui<sup>1,6</sup>, S. Lohse<sup>1,6</sup>, N. Martynova<sup>4</sup>,  
E. Minaya Ramirez<sup>11</sup>, A. Mistry<sup>1,2</sup>, T. Murboeck<sup>1,2</sup>, Yu. N. Novikov<sup>4,7</sup>, S. Raeder<sup>1,2</sup>, D. Rodriguez<sup>10</sup>,  
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One of the fundamental questions in nuclear physics concerns the existence and properties of the heaviest elements. To solve such a puzzle, it is essential to investigate the nuclear shell effects that counteract spontaneous fission and determine the stability of such exotic systems.

High-precision Penning-trap mass spectrometry (PTMS) is an established tool for investigations of shell effects and their evolution for nuclear systems with different proton to neutron ratios through direct measurements of the atomic masses and hence binding energies [1]. The heaviest elements investigated to date in pioneering experiments with the SHIPTRAP setup at GSI, have been nobelium and lawrencium [2,3]. The direct measurement of the masses of <sup>252–255</sup>No and <sup>255,256</sup>Lr has allowed mapping the strength of the deformed subshell closure at  $N=152$ .

Recent developments of the setup allowed pushing these limits to even heavier and more exotic nuclei in the latest beam time at GSI (June-July 2018) when the mass of the first super-heavy element, <sup>257</sup>Rf ( $Z=104$ ) was directly measured for the first time. The SHIPTRAP efficiency has been boosted by the implementation of a cryogenic gas-catcher with increased stopping and extraction ion efficiency [4]. The mass resolving power, precision and detection sensitivity has been further enhanced by the development, at SHIPTRAP, of the Phase-Imaging Ion-Cyclotron-Resonance technique [5,6]. This state-of-the-art method allowed also, during the same experiment, to simultaneously measure ground and low-lying isomeric states of the heaviest elements, which are often not accessible by other techniques.

The setup upgrade as well as the latest results and the related new piece of information concerning the shell structure of the heaviest elements will be discussed together with the future plans.

## References

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## Adsorption of the Superheavy Element Species on Gold Surface: Relativistic Density Functional Study

Yu. Demidov

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Long-lived isotopes of the superheavy elements (SHE) with atomic numbers  $Z \geq 104$ , can be produced in fusion reactions between heavy actinide targets and neutron-rich projectiles at only very low rates: from single atoms per minute ( $Z = 104$ ) to single atoms per week ( $Z = 114$ ). Since the thermochromatography on gold has proved a unique method for chemical detection of heaviest elements, the description of SHE – gold interactions has recently been of prime concern. It has been shown experimentally that the adsorption energies of Cn ( $Z = 112$ ) and Fl ( $Z = 114$ ) atoms on gold surface are close and lower than those for their closest homologues Hg and Pb, respectively. This confirms the theoretical predictions concerning the electronic structure of the Cn and Fl atoms: due to strong relativistic stabilization of  $s$  and  $p_{1/2}$  shells, both Cn ( $6d^{10}7s^2$ ) and Fl ( $6d^{10}7s^27p_{1/2}^2$ ) ground states are of closed-shell character. Yet, positive chemical identification of Fl and its separation from Cn remains problematic. To date, two experiments lack reproducibility and provide conflicting data on Fl adsorption behavior on gold surface.

A substantial difference in the high-level theoretical predictions of the ionization potentials of Cn (1155 kJ/mol) and Fl (824 kJ/mol) supports possible chemical dissimilarity of these elements. While the results of thermochromatography on gold remain inconclusive, chemical separation of Cn and Fl can become feasible with a more reactive surface. Recent experimental efforts have been focused on selenium as possible adsorbent and model experiments based on the adsorption of Hg, a lighter homologue of Cn. First attempt to provide estimates of the adsorption energy of Cn or Fl on a selenium surface based on Hg-, Cn-, and Fl-Se<sub>n</sub> cluster models of the adsorption complexes is reported.

Strong relativistic effects suggest dramatic dissimilarities in the chemical behavior of SHEs and their formal lighter homologues (see e.g. [1]). The calculated adsorption energy for single atoms of nihonium on a gold surface differs substantially from the experimentally measured adsorption energy on gold of its nearest homolog, thallium. This casts doubt on the usefulness of the experiments with Nh formal homologues for understanding its chemistry. Despite manifest deviations of the chemical properties of the SHEs from the trends observed in their lighter formal homologues in the respective groups of the periodic table, finding chemical pseudo-homologues appears a practically meaningful issue.

Due to this unique feature of the 7<sup>th</sup> row of the Periodic Table, the electronic structure of a Nh atom can be interpreted as a Fl atom with a hole in its closed  $7p_{1/2}$ -subshell. This observation seems to render astatine a closer chemical “relative” of Nh in comparison to the formal homologue Tl. Thus, At might be a plausible chemical species for model experiments aiming at finding the optimum experimental conditions for further explorations of the Nh chemistry. The predicted adsorption energies for At & AtOH on gold are  $130 \pm 10$  kJ/mol and  $90 \pm 10$  kJ/mol, respectively [2]. This confirms the experimental observation on the formation of AtOH molecules in presence of trace amounts of water and oxygen in the carrier gas. Due to the similarities in the chemical properties of AtOH and NhOH molecules, one may expect, that the formation of NhOH is indeed possible, under experimental conditions similar to those in the experiments on At.

### References

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## Optimizing the in-situ production yield of transition metal carbonyls

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Despite the first synthesis of Sg(CO)<sub>6</sub> in 2014 by Even et al. being successful [1], also further experiments performed at RIKEN targeted at measuring thermodynamic properties of this new compound suffered from low formation rates for carbonyl complexes in general. In the mentioned measurement campaign performed in 2016, the post-separator yield was in the range of about 5-10% for Sg(CO)<sub>6</sub>, the same for W(CO)<sub>6</sub> and only slightly higher (20%) for Mo(CO)<sub>6</sub>, making the experiment analysis difficult due to low statistics for the observed number of events for the transactinide.

In order to overcome this problem, this work focuses on increasing the post-separator yield of Mo(CO)<sub>6</sub> in a model system called FORA, based on a down-scaled version of the RIKEN-setup used in 2016. The studied, short lived Mo-Isotopes are generated by the spontaneous fission process of a <sup>252</sup>Cf source and the yield of metal-carbonyls produced under various reaction conditions is monitored by trapping these volatile compounds on a charcoal-trap and monitoring their decay using  $\gamma$ -spectroscopy. Since additionally to Mo-Isotopes, the fission of <sup>252</sup>Cf also produces significant amounts of Tc, Ru and Rh, it was decided to extend our studies to those elements as well. A simple model was developed, that allows to calculate an absolute post-separator chemical yield for the synthesis of Mo(CO)<sub>6</sub>, including a correction for the transportation time, that might differ between the model-system used here and an actual accelerator-based setup.

Using the mentioned setup, the dependency of the carbonyl-formation reaction on various parameters was investigated. Those parameters include pressure, gas mixture composition, kinetic energies of the recoiling isotopes, gas velocity, various impurities including H<sub>2</sub>, O<sub>2</sub> and CH<sub>4</sub> as well as different purification columns and getters used to clean the applied gas mixtures. The results of those investigations will be presented. Further studies including the influence of temperature and H<sub>2</sub>O will be performed in the near future.

### References

- [1] J. Even et al., Science, **345**, 6203 (2014)

## Beam commissioning of cw Linac demonstrator

M. Miski-Oglu

*Helmholtz Institute Mainz, Germany*

A newly developed superconducting 15-gap RF-cavity has been successfully tested at GSI Helmholtzzentrum für Schwerionenforschung. After a short commissioning and ramp up time of some days, a Crossbar H-cavity accelerated first time heavy ion beams with full transmission up to the design beam energy of 1.85 MeV/u. The design acceleration gain of 3.5 MV inside a length of less than 70 cm has been verified with heavy ion beam of up to 1.5 particle  $\mu$ A. The measured beam parameters showed excellent beam quality, while a dedicated beam dynamics layout provides beam energy variation between 1.2 and 2.2 MeV/u. The beam commissioning is a milestone of the R&D work of Helmholtz Institute Mainz and GSI in collaboration with Goethe University Frankfurt towards a superconducting heavy ion continuous wave linear accelerator cw-Linac with variable beam energy. Further linac beam dynamics layout issues will be presented as well.



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## Commissioning of the recoil separator MARA at JYFL and the first experiments

J. Sarén, J. Uusitalo and the Nuclear Spectroscopy Group

*Physics Department, University of Jyväskylä, Jyväskylä, Finland*

MARA is a new in-flight recoil separator mainly for studies of the exotic isotopes close to the N~Z line and close to the proton-drip line produced in fusion evaporation reactions. MARA consists of a quadrupole triplet followed by an electrostatic deflector and a magnetic dipole. It has two main functions: separate a primary beam from products and give additional mass over charge selectivity among the products. The mass resolving power is roughly from 100 to 300 depending on the experiment. MARA is a complementary device to the gas-filled RITU separator and enables the continuation of the nuclear spectroscopy research program in Jyväskylä to lighter masses.

In this presentation basic properties of MARA and its auxiliary detector setups will be presented. The commissioning campaign and the accomplished scientific experiments and their highlights, mainly the variety of new isotopes, will be reviewed and an overview of the experimental conditions based on the reactions performed with MARA will be given. Some of the physics cases will be discussed alongside the spectrum of the accepted proposals.

## Target development for $S^3$

Ch. Stodel and the  $S^3$  collaboration

*GANIL, Caen, France*

In the first phase of exploitation of SPIRAL-2, the LINAC beams will be delivered to the NFS (Neutrons For Science) and  $S^3$  (Super Separator Spectrometer) experimental halls. These facilities were designed according to the Letters of Intent submitted by a large physics community (SPIRAL2 Letters of Intent). With  $S^3$ , special emphasis is on the study of rare nuclei, such as superheavy elements and neutron-deficient isotopes, produced by fusion evaporation reactions. The spectrometer includes a rotating target (Stodel, et al., 2015) (Kallunkathariyil, et al., 2018) to sustain the highly intense heavy ions beams, a two-stage separator (momentum achromat followed by a mass spectrometer (Dechery, et al., 2016)) that can be coupled to the implantation-decay station SIRIUS or to a gas catcher (Piot, 2012).

In order to cope with the very low production rate of the rare events, the beam intensities will be higher by a factor of five to ten compared to the present ones. Then, a major experimental concern is the behavior of thin targets under these highly intense heavy ion beams.

We propose to report on the target's stations including their instrumentation and on the envisaged target developments.

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- [2] C. Stodel et al, «High Intensity targets stations for  $S^3$ ,». J. Radioanal. Nucl. Chem. 305, 761-767 (2015).
- [3] J. Kallunkathariyil et al, « $S^3$  Target Monitoring with an Electron gun,» AIP Conference Proceedings, to be published, 2018.
- [4] F. Dechery et al, «The Super Separator Spectrometer  $S^3$  and the associated detection systems: SIRIUS & LEB-REGLIS3». Nucl. Inst. Meth. B. 376, 125-130 (2016).
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## Central depression of nucleonic densities

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The densities of atomic nuclei are an intriguing and important subject of current research. In most nuclei the density distribution reaches nuclear saturation density in the center and smoothly decreases to zero in a surface region. However, in superheavy nuclei the Coulomb repulsion between protons in the center becomes so strong that an appreciable depression in the central density results. Our nuclear density functional theory (DFT) calculations indicate that the so-called nuclear semi-bubbles are already visible in elements heavier than Pb and the newest addition to the periodic table, Og, presents already a strong central proton density depression.

The consequences of central density depression are many-sided. High nucleon  $j$ -orbits become lower in energy, low- $j$  orbits become higher. On the other hand rms radii become much larger. Our results show that isotopic shifts in Nobelium isotopes can be well reproduced with the central depression.

Work supported by: U.S. Department of Energy DOE-DE-NA0002847 (NNSA, the Stewardship Science Academic Alliances program), de-sc0013365 (Office of Science), de-sc0008511 (Office of Science, NUCLEI SciDAC-3 collaboration) and BMBF-Verbundforschungsprojekt (05P15RDFN1).



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Recoil Separator for Superheavy Element Chemistry

## Rapid extraction of short-lived isotopes from a buffer gas cell for use in gas-phase chemistry to access elements beyond Fl

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In recent years, the chemical properties of the SuperHeavy Elements (SHE) copernicium (Cn,  $Z = 112$ ) and flerovium (Fl,  $Z = 114$ ) and their homologs have been extensively studied [1,2]. One main motivation for performing such experiments is to elucidate the influence of relativistic effects on chemical properties of these elements [3]. Due to low production rates and short half-lives, only single atoms are available in chemical experiments. The combination of gas-phase chromatography setups with an electromagnetic preseparator like TASCA proved to be the best experimental approach for reaching the required sensitivity for atom-at-a-time chemical studies [4]. After production via fusion-evaporation reactions and separation in, e.g., TASCA, these atoms are thermalized in the gas-filled volume and flushed to the chromatography setup with a rapidly flowing gas. This approach is currently applicable to isotopes with half-lives longer than about 0.5 seconds, and is limited by the extraction time of hundreds of milliseconds for recoils under such conditions. For elements beyond Fl, half-lives of suitable isotopes drop significantly below that level. The probably most suitable isotope of moscovium (Mc,  $Z = 115$ ),  $^{288}\text{Mc}$ , which is accessible directly via the  $^{48}\text{Ca} + ^{243}\text{Am}$  reaction with a comparatively high cross section of about 10 pb, and has a half-life of  $164^{+30}_{-21}$  ms [5]. To overcome this limitation, exploratory experiments were carried out with the aim to test a potentially faster system comprising the gas phase chromatography setup COMPACT [3] coupled to an existing buffer gas stopping cell operated with electric fields [5]. In this contribution, an overview of achieved performance, further optimization of the system, and the development of a new Mini-COMPACT-design will be discussed.

## References

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