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Production and properties of the heaviest elements

Status and perspectives

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Abstract. This article reviews the following topics which were discussed at the 375th Wilhelm and Else Heraeus-Seminar *Workshop on the Atomic Properties of the Heaviest Elements* held from September 25–27, 2006 at the Abtei Frauenwörth im Chiemsee, Germany: (i) the recent progress in the production of the heaviest elements, the investigation of their nuclear structure, and prospects for direct mass measurements in Penning traps. (ii) Recent studies of their chemical properties with the aid of volatile species and single-atom aqueous-phase chemistry; (iii) the current status and future prospects for the investigation of atomic and ionic properties such as optical spectroscopy in gas cells and ion traps, including fully relativistic calculations of the atomic level structure with predictions for the element nobelium; and (iv) ionic charge radii measurements in buffer gas filled drift cells, and ion chemical reactions in the gas phase.

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1 Introduction

Trans-fermium elements with charge numbers $Z > 100$ can only be produced by nuclear fusion reactions with very low rates. For the heaviest elements the production rate may be as low as only one atom per month (or even less). Already their identification is a real challenge, all the more the investigation of their properties. At the same time, production of superheavy elements and investigation of their properties is a multidisciplinary field of research which comprises experimental and theoretical nuclear physics, single-atom chemistry, quantum chemistry with state-of-the-art computational methods, as well as experimental and theoretical atomic physics. The 375th Wilhelm and Else Heraeus-Seminar *Workshop on the Atomic Properties of the Heaviest Elements*, which was held from September 25–27, 2006 at the Abtei Frauenwörth im Chiemsee, Germany, brought together experts in the above mentioned fields. Their contributions are collected in this topical issue of EPJ D from which the strong interconnection of the various fields becomes apparent. It ranges from questions concerning efficient nuclear fusion reactions over relativistic and quantum electro dynamical effects to the chemical and atomic properties of superheavy elements. The aim of this article is

not only to summarize the workshop but also to introduce a broader audience into this intriguing field of superheavy element research with all its facets. In analogy to the workshop schedule, the paper is organized into the following sections: "Production of superheavy elements and nuclear properties", "Chemical properties", "Atomic and ionic level structure", "Ion chemical reactions and ion mobility in buffer gases" and "Conclusion".

2 Production of superheavy elements and nuclear properties

Synthesis of superheavy elements (SHE) and studies of their nuclear properties can be divided into the categories: production, nuclear structure investigations, mass measurements, and nuclear theory which will be discussed in the following.

2.1 Superheavy element production

Technically, investigations of nuclear fusion reactions possibly suited to produce isotopes of so far unknown elements and to identify them on the basis of very few observed radioactive decays, are conducted in several steps.

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The first one, also applied in nuclear structure investigations, is to separate the products from complete fusion reactions in-flight from the primary beam and to implant them into silicon detector arrangements placed in the focal plane of the separator. Different types have been used in the experiments presented: the gas-filled separators DGFRS (FNLR-JINR Dubna) [1], GARIS (Riken, Tokai) [2], RITU (University of Jyväskylä) [3] and BGS (LNBL, Berkeley) [4] as well as the velocity filter SHIP [5] at GSI, Darmstadt. Discrimination of eventually superheavy nuclei from 'residual' background is achieved by establishing delayed coincidences between implanted nuclei and succeeding α decays or spontaneous fission. Commonly such sequences are denoted as 'correlation chains' or 'decay chains'. If at least one α decay having properties (energy, life-time) in-line with that of a known nuclide, new isotopes forming the heads of such chains can be identified safely. If this is not the case additional measures are required: the comparison of measured α-decay energies and life-times with systematics (Geiger-Nuttal-rule) can be used to establish the atomic number of the produced nucleus. Also from the production mechanism itself information on mass and atomic number of the evaporation residue can be inferred i.e. by the determination of the production cross section as function of the bombarding energy (excitation functions). In addition, so-called cross bombardments are pursued. This method implies to measure the production cross sections for a specific isotope in dependence of target projectile combinations leading to compound nuclei slightly differing in mass and atomic numbers, which then often allows to determine the most probable mass and atomic number of the unknown isotope, or at least sets limits to their values.

Two types of projectile-target combinations have been applied so far to produce superheavy elements: the first one is the use of target nuclei around doubly magic ^{208}Pb and medium heavy projectiles as ^{64}Ni or ^{70}Zn as presented in an overview given by one of the authors (F.P.H.). The advantage of this combination are low excitation energies ($E^* < 20$ MeV) of the compound nuclei at the fusion barrier. New elements with atomic numbers $Z = (107-112)$ have been first produced at SHIP by this method. It was found that cross sections typically decrease by a factor of ≈ 3.5 per increasing atomic number. Morita [6] presented results from experiments performed at GARIS which aimed at the production of still heavier nuclei using 'cold' fusion reactions. The isotope ^{278}113 was synthesized in bombardments of ^{209}Bi with ^{70}Zn. In several experimental runs covering in total 241 days of irradiation, two decay chains of ^{278}113 were observed, which resulted in a production cross section of only ≈ 30 fb.

The other way to produce superheavy elements is the use of doubly magic ^{48}Ca as projectiles and actinide as targets. These combinations result in higher excitation energies of the compound nuclei but complete fusion is believed to be less hindered than in the more symmetric reactions using targets of elements around ^{208}Pb. These reactions have been consequently applied at DGFRS during the past decade and the production and identification

of nuclides attributed to isotopes of elements up to $Z = 118$ has been reported by Yu.Ts. Oganessian [7]. The production cross sections remained quite constant in the range $Z = (112-118)$, which was regarded as a signature of enhanced nuclear stability with increasing proton and neutron numbers and thus as a signature for approaching the predicted spherical 'superheavy shells' at $Z = 114$ and $N = 184$. So far the published data are convincing, but their interpretation, i.e. their assignment to the decay of isotopes of elements $Z \geq 112$ is still under discussion.

Thus an examination of the Dubna results seemed necessary. As a first step attempts to produce and to identify the isotope ^{283}112 using the projectile-target combination ^{48}Ca + ^{238}U were performed at BGS and at SHIP. For the latter isotope disagreeing decay properties were obtained at the energy filter VASSILISSA (FNLR-JINR Dubna) and the DGFRS. The experiments at BGS and first experiments at SHIP could reproduce neither of the Dubna results. With respect to this situation, P. Armbruster considered pre-compound charged particle emission as an alternative to interpret the results from the ^{48}Ca induced reactions on actinide targets which are reported to yield e.g. element 118 [7]. The combination of velocity filters or gas filled separators with fast chemical analysis will allow the mandatory identification of nuclear charge number Z and mass number A, needed for the identification of the multitude of isotopes producible by this mechanism.

In an irradiation of ^{238}U with ^{48}Ca performed at SHIP in January/February 2007 [8], two decay chains were observed each consisting of implantation of a 'heavy' nucleus, followed by an α decay and finally terminated by a spontaneous fission event. The α energy and life-times are in agreement with the data attributed in the Dubna-DGFRS experiments as presented by Yu.Ts. Oganessian to ^{283}112 and ^{279}110. Since evaporation residues from complete fusion reactions have kinematic properties (velocity, angular distribution) different to those from pre-compound reactions, the latter should be strongly suppressed by SHIP. P. Armbruster explains in this volume [9] how these new findings render pre-compound charged particle emission seemingly of no concern for the production of the isotopes observed previously in reactions of ^{48}Ca with actinide targets at the DGFRS [10,11].

Obviously, all experiments suffer from low numbers of events that can be observed in reasonable irradiation times. A higher sensitivity is needed, which calls for upgrade of accelerators, development of targets that can withstand significantly higher beam currents, and improvements of separators to increase efficiency and suppression of the primary beam and products from other nuclear reactions than complete fusion.

2.2 Nuclear structure investigations

Significant progress has been achieved in the past decade in the field of nuclear structure investigations by use of detector setups with high sensitivity and energy resolution. One major tool is the combination of in-beam γ and conversion electron spectroscopy with in-flight separation

of the complete fusion products from the projectile beam. Evaporation residues registered and/or identified by their radioactive decay in the focal plane of the separator are then used to tag γ decays or conversion electrons registered with detectors placed around the target. Thus, γ rays or electrons emitted during deexcitation of the compound nucleus can be discriminated from the huge background of radiation stemming from other nuclear reactions and prompt fission of the compound nuclei. Using this method, mainly at the RITU separator at the University of Jyväskylä, ground-state rotational bands in several even-even nuclei (^{254,252}No, ^{250,248}Fm) up to spins $22\hbar$ and also rotational bands in odd-mass nuclei (^{253}No, ^{255}Lr and ^{251}Md) have been investigated. An overview of these results was given by R.-D. Herzberg [12,13].

The second focus of nuclear structure investigations is on decay spectroscopy in the focal plane after in-flight separation. One of us (F.P.H.) presented an overview [14] of the results obtained at SHIP during the last years. One field of this research subject are α-γ- or α-conversion electron (CE) decay studies. Since higher levels are in general only weakly populated by α decay, such experiments deliver information on nuclear structure essentially at excitation energies below 500 keV. Since in odd-mass nuclei nuclear structure at low excitation energies is mainly governed by the unpaired nucleon, so far investigations were focussed on this subject. Systematic studies of low-lying Nilsson levels were performed for $N = (145-151)$ isotones up to $Z = 104$ and for odd-mass einsteinium isotopes at SHIP. Another feature of spectroscopy in the focal plane are decay studies of isomeric states, mainly performed at RITU and at SHIP. Gamma decay patterns of K-isomers in ^{254}No and ^{250}Fm, for which so far only indirect evidence was existing, were measured for the first time. In addition, new isomeric states in $^{251-254}$No were discovered. Their half-lives and decay properties proved a high stability of these nuclei at high spins and gave new insights on properties of single particle levels in deformed superheavy nuclei. For the future, precise spectroscopy of mass separated superheavy nuclei and investigation of spontaneous fission are envisaged.

Chemical separation was shown to provide a link between nuclear spectroscopy and investigation of chemical properties, since this technique provides clean samples well suited for measuring decay properties especially in the case of low production rates. Recent results [15] obtained for hassium isotopes produced in the reaction ^{248}Cm(^{26}Mg, xn) $^{274-x}$Hs were presented by J. Dvorak.

Finally, in context with the latest results on superheavy element production and their controversial discussion at the symposium, nuclear reaction studies were resumed. M. Morjean [16] presented data from experiments performed at INDRA-GANIL where reaction times for the system ^{238}U + Ge at 6.1 AMeV were investigated. As in previous experiments using the projectile-target combination ^{238}U + Ni, also here lifetimes $> 10^{-18}$ s of the composite system were observed, which are interpreted as an indication that the corresponding compound nuclei with atomic numbers $Z = 124$ and $Z = 120$ may

have high fission barriers. Massive transfer in head-on collisions of ^{25}Mg on ^{206}Pb has been investigated at SHIP. Transfer of carbon clusters on the target nucleus was observed. Although this result is interesting its relevance for the production of heaviest nuclei has to be proven.

As a general trend, nuclear structure investigations are a growing field and besides development of more refined detector systems new setups will be available in future: SAGE, a spectrometer suited for simultaneous in-beam conversion electron and γ spectroscopy will be installed at RITU. The status of the project was presented by R.-D. Herzberg. TASCA, a gas-filled separator with high efficiency for products from very asymmetric reactions, e.g. ^{22}Ne + ^{238}U, is presently commissioned at GSI and will in future, besides for chemistry experiments, also be used for nuclear structure investigations complementary to SHIP as presented by M. Schädel [17]. Technical details on TASCA were explained in a poster by A. Semchenkov.

2.3 Mass measurements

Until now the knowledge of masses in the SHE region depends mostly on the interconnection of masses via α-decay measurements, thus with errors that accumulate progressively. Moreover, if the decay chain ends in a fissile nuclide these masses cannot be anchored to known masses. Thus, direct mass measurements will significantly attribute to the determination of the nuclear masses of superheavy elements and their mass number assignment. Moreover, they may help to disentangle subsequent α-decays from isomeric states.

Within the past 17 years Penning trap mass spectrometers have evolved into the most precise tool for mass measurements of exotic nuclides. ISOLTRAP, the first tandem Penning trap mass spectrometer has been developed at ISOLDE, CERN [18–20]. Today, its precision amounts to $\delta m/m \approx 10^{-8}$ [21]. The unique possibilities existing now by the coupling of SHIP to such a Penning trap for mass measurements required a suitable interface the development of which has a history as long as the development of the mass spectrometer itself: already 20 years ago first concepts [22] were developed based on buffer gas stopping cells and mass selective ion detection for the investigation of fusion reaction products. Singly charged ions come to rest in a helium buffer gas pressure of about 1 bar. In the so-called Ion Guide Isotope Separation On-Line (IGISOL) [23,24] technique, the ions are flushed out by the gas stream through a nozzle. Subsequently the ions are separated from the buffer gas in differential pumping sections, mass separated and delivered as low-energetic radioactive beams to nuclear mass measurements or collinear laser spectroscopy setups [25]. The combination of buffer gas stopping and electric field guidance of the ions result in extraction times in the order of 1 ms at rather low gas flows. Such a system has first been described in reference [26]. Succeeding setups [27–32] use additional RF-potentials at the electrodes which repel the ions from the walls of the buffer gas cell similar to the

ion catcher setup developed at the tandem accelerator facility of the Maier-Leibnitz-Laboratory in Munich, which was presented by Jürgen Neumayr [33].

Also at SHIP the transition is made by stopping the fusion products in a He-filled gas cell [28] and extraction of the charged fraction through a nozzle. Aims in the near future are mass measurements with this SHIPTRAP facility in the superheavy element region around ^{254}No. The feasibility of such direct mass measurements has been demonstrated by M. Block in his contribution on the example of measuring the masses of very proton-rich evaporation residues in the range $A \approx (90-160)$ [34]. Technical aspects of SHIPTRAP [35] were presented by A. Chaudhuri and A. Martin in the poster session.

2.4 Nuclear theory

Relativistic Hartree-Bogoliubov (RHB) [36] theory is used to calculate energy surfaces and Q_α-values for chains of superheavy elements. P. Ring presented a new covariant density functional derived from high precision fits to experimental data which reproduces nuclear binding energies with an uncertainty of $\delta m = 900$ keV throughout the whole periodic table [37]. This reflects in the high accuracy of $\delta m/m = 5 \times 10^{-4}$ for superheavy elements and leads to a convincing reproduction of the Q_α values [38] derived from the decay chains of element 115 [39]. Calculations of the shell structure of superheavy nuclei by these self-consistent mean-field models yield $Z = 120$ and $N = 172$ for the next magic proton and neutron numbers beyond $Z = 82$ and $N = 126$ of ^{208}Pb. In the region around $^{292}_{172}120$ a deep central depression of nuclear density is predicted — a striking phenomenon often referred to as "Hollow"- or "Bubble"-nuclei [40].

Production cross sections of the heaviest nuclei in complete fusion reactions are discussed by R.N. Sagaidak within this volume [41]. Systematics in the fusion probability as function of the fissility are given which can be used to choose the most effective reaction for the synthesis of unknown nuclei.

3 Chemical properties

Nuclear chemists usually associate the term 'superheavy element' with 'transactinide element' meaning that the vanishing of the liquid drop fission barrier coincides with the completion of the actinide series. Therefore, the superheavy elements begin with the element rutherfordium ($Z = 104$). Chemical investigations of superheavy elements are experimentally very challenging, but feasible, as will be outlined. Usually, in a first step, the group properties of a transactinide element are determined followed by more sophisticated experiments to elaborate more subtle chemical differences to the lighter homologues. Besides the experimental challenges, the chemical properties of transactinides are of interest due to the expected strong influence of relativistic effects. Therefore, predictions about the chemical behavior of transactinide elements are pivotal.

There are two approaches to predict chemical properties of superheavy elements. The common approach is to search for empirical relationships of i.e. thermochemical properties that undoubtedly exist within the groups and periods of the periodic system and to extrapolate these properties to the transactinide elements. Such an approach neglects the fact that relativistic effects roughly increase proportional to Z^2. Nevertheless, such predictions have proven valuable in designing experiments with transactinide elements. The inclusion of relativity is, scientifically speaking, the only correct way to proceed. However, there are limits to what theoretical chemists are able to calculate.

One aspect, that should not be neglected, is, that also nuclear chemists currently rely on the detection of the nuclear decay properties of a chemically isolated nucleus in order to prove, that indeed a transactinide element was isolated and studied. In this respect nuclear chemists also contribute valuable nuclear decay data to the field of superheavy element research. In special cases, chemical separation schemes proved to be superior to separation by on-line kinematic separators and resulted in the discovery of new isotopes [15,42] or the confirmation of previously reported decay properties [43].

3.1 Relativistic effects on atomic and molecular properties of transactinides

Commonly, three relativistic effects on the atomic properties of heaviest elements are being distinguished. The contraction and stabilization of the spherical s and $p_{1/2}$ orbitals are considered as the "direct" relativistic effect. Although this effect is strongest for the inner K and L electrons, it is still large for the outermost s and $p_{1/2}$ orbitals, leading to a 25% contraction of the radial distribution of $7s$ electrons in dubnium. The relativistic contraction leads to a more efficient screening of the nuclear charge and, as a consequence, to an expansion and an energetic destabilization of d and f electron orbitals. This effect is known as "indirect relativistic effect" and is important for orbitals which are involved in chemical bonding. The third relativistic effect concerns the "spin-orbit splitting" of levels with $l > 0$, which in transactinide compounds becomes similar or even larger in size than typical bond energies. Keeping in mind that all three relativistic effects contribute by about the same order of magnitude, the prediction of the chemical behavior of transactinides and their compounds is by no means an easy task.

In her contribution, V. Pershina [44] focused on the chemical properties of element 112 (E112), one of the most interesting transactinide elements. E112 is expected to exhibit the strongest relativistic effects of all known transactinide elements. As an analog to the group 12 element Hg, E112 is expected to be relatively inert and volatile. Early considerations even speculated about the possibility of E112 behaving like a noble gas [45]. Calculations were performed using Dirac-Fock (DC), 4-component Density Functional Theory (4c-DFT) and spin polarized 4c-DFT. These theoretical investigations concerning atomic properties as well as molecular properties of diatomic compounds

and cluster calculations of M–Au$_n$ (M = Hg, E112) clusters, painted a more diverse picture of the chemical properties of E112. On inert surfaces such as quartz, E112 is expected to interact stronger than Hg due to the relativistically contracted atomic radius. Upon adsorption of E112 on Au clusters, the influence of relativistic effects depends on the adsorption position (on-top, hollow, or bridge). In the hollow position, relativistic effects increase the interaction energy of E112 with the Au cluster, while "non-relativistically" E112 would be more volatile. Nevertheless, also relativistic E112 should be more volatile on a Au(100) surface than Hg, but far from being as inert as i.e. a noble gas.

3.2 Volatile transactinides and transactinde compounds

In the last couple of years gas-phase chemistry experiments with transactinide elements have proven extremely successful. Early on, volatile halide and oxyhalide complexes were investigated. This technique dates back to the early sixties and has been used by I. Zvara and co-workers to start up chemical experiments with transactinide elements in Dubna (see Refs. [46,47] for reviews). In the mean time the volatility of RfCl$_4$ [48] and RfBr$_4$ [49] in comparison to their lighter homologues has been investigated. Db seems to be more sensitive to oxygen and rather forms DbOBr$_3$ or DbOCl$_3$ than the pure halides [50,51]. Element 106, seaborgium, was shown to form volatile SgO$_2$Cl$_2$ [52], whereas element 107, bohrium, was isolated for the first time as BhO$_3$Cl [53]. In all cases a thermochemical property, namely the enthalpy of adsorption on the chromatographic surface, usually quartz, could be derived, even though in most cases only few atoms have been registered. Some transactinide elements form volatile oxide-hydroxides and Sg was shown to exhibit this property as well [54]. In his review about superheavy element chemistry experiments performed at GSI, M. Schädel discussed aqueous and gas-phase chemistry experiments [47]. One of the highlights concerned the high volatility of group 8 oxides. Thus, hassium was isolated with very high yield as HsO$_4$ [55]. This extremly powerful technique opened chemists the door to measure picobarn and subpicobarn cross sections and eventually led to the discovery of the new doubly magic nucleus ^{270}Hs [15]. HsO$_4$ was shown to react with NaOH and to form the disodium-tetraoxohassat(VIII) Na$_2$[HsO$_4$(OH)$_2$], similar to its lighter homolog Os [56].

One of the most fascinating new experiments was reported by H.W. Gäggeler and co-workers [43], which after a series of unsuccessful attempts reported a breakthrough in chemical investigations of E112. Fusion products of the reaction ^{48}Ca + ^{242}Pu were thermalized in dry He gas, which flowed to a thermogradient detector with gold-covered detector surfaces. Two α-spontaneous fission decay chains were registered in the warm section of the thermogradient, where also short-lived Hg-isotopes were deposited. Radon isotopes were registered near the exit at the cold end of the detector. The two decay chains were

attributed to the α-particle decay of the nuclide ^{283}112 followed within several hundred milliseconds by spontaneous fission of ^{279}Ds. The same decay properties have been observed previously at the DGFRS in the reaction ^{48}Ca + ^{238}U, and as terminating members of decay chains observed in the reaction ^{48}Ca + ^{242}Pu [57]. This result is significant in two ways. First, E112 seems to behave chemically more like Hg as it interacts strongly with Au. Second, it appears as if indeed the formation of a superheavy element in ^{48}Ca-induced reactions on a actinide target was confirmed using a chemical separation technique. This finding opens up a new area of superheavy element chemistry. In his poster presentation, A. Yakushev presented first test experiments with short-lived α-decaying Pb isotopes. The proposed experiment focuses on the question whether element 114 (E114) is more or less inert than Pb. The idea is to use a hot metal catcher to adsorb Pb-like, but not the more volatile Hg-like products. If E114 is more inert and thus more volatile than Pb, it would pass the hot metal catcher and be registered in a thermogradient detector. If E114 is adsorbed, its α-decay daughter, E112, would desorb from the hot metal catcher and be registered in the detector.

3.3 Aqueous phase chemistry with tansactinides

Early on, aqueous chemistry experiments played and still play an important role in transactinide chemistry. Aqueous phase chemistry usually offers very good separation factors and thus delivers clean samples to α-particle spectroscopy. This, however, at the cost of speed and overall efficiency. In his contribution, M. Schädel introduced the automated rapid chemistry apparatus ARCA, which has been successfully used to investigate the chemical properties of Rf, Db, and Sg using liquid chromatographic techniques (see Refs. [47,58] for reviews). In a first experiment [59] Sg was shown to elute as hexavalent ion from a cation exchanger in 0.1 M HNO$_3$/5 × 10^{-4} M HF. Over 5000 column separations were needed to detect 3 α-α correlations from the decay of ^{261}Rf and ^{257}No daughters of ^{265}Sg. From this experiment it was concluded that Sg was eluted as neutral of anionic complex i.e. SgO$_2$F$_2$ or SgO$_2$F$_3^-$, similar to the lighter homologs Mo and W, and not like the pseudo-homolog U (no SgO$_2^{2+}$). A second experiment [60] performed in pure HNO$_3$ [47] showed no formation of neutral or anionic complexes of Sg i.e. no SgO$_4^{2-}$ in contrast to W. This was attributed to the decreasing hydrolysis in the series Mo > W > Sg. An improved version of ARCA was built at JAEA and named AIDA. In this apparatus the manual transport of the final samples from the drying position to the detector chambers was also automated. This system has been used to investigate the ion exchange properties of Rf in detail. In his contribution, A. Toyoshima presented the results of fluoride complexation studies of Rf. In previous studies the anion exchange behavior of Rf in 1.9–13.9 M HF has been studied and was found to be quite different than that of the group 4 homologs Zr and Hf [61]. To further understand the fluoride complexation of Rf the anion exchange behavior from mixed HF/HNO$_3$

solutions was studied. It was found that the formation of the hexafluoro complex of Rf, $[RfF_6]^{2-}$, is much weaker than those of $[ZrF_6]^{2-}$ and $[HfF_6]^{2-}$.

3.4 New developments in chemical investigations of transactinides

For chemical investigations, the element of interest has to be transported rapidly from the location of its production to a chemistry laboratory, or, the element has to be converted in-situ to a chemical compound. Both approaches have a number of drawbacks. Usually, the element of interest is transported attached to small aerosol particles in gas through a capillary to the chemistry laboratory. This transport can be accomplished within few seconds. Unfortunately, the transport efficiency is negatively affected by increasing beam intensities. The transport is unspecific, thus also undesired by-products of the nuclear reaction are transported. The harsh ionizing conditions created by the beam passing through the target recoil chamber are severely limiting the in-situ synthesis of volatile transactinide compounds. Therefore, the use of an efficient kinematic pre-separator was considered. First experiments with so-called recoil transfer chambers behind gas-filled separators immediately proved very successful, due to the suppression of the intense primary beam and the strong suppression of undesired by-products of the nuclear reaction. J.P. Omtvedt [62] reported about liquid-liquid extraction experiments with ^{257}Rf using the Berkeley Gas-filled Separator BGS as a pre-separator. Since the so-called SISAK technique, relies on liquid scintillation counting as detection method the use of a pre-separator was essential. An extraction system to study Db with the SISAK system was developed and presented by J. Gates. In on-line and off-line studies the extraction of Nb and Ta from 1–11 M HCl/LiCl into 0.01 M di-(2-ethylhexyl)-phosphoric acid (HDEHP) in chloroform was studied. Significant differences in the extraction behavior of Nb and Ta were observed from HCl(1M)/LiCl solutions with HDEHP. At a chloride concentration of 11 M Ta is extracted quantitatively while Nb extracts to a much lesser extent. At RIKEN a recoil transfer chamber was successfully tested behind the gas-filled separator GARIS. The impressive results about the startup of superheavy element chemistry at RIKEN, Japan, were presented by H. Haba [63]. At GSI a dedicated gas-filled separator TASCA is currently being commissioned. Using the magnets of the old HECK separator, A. Semchenkov in his poster presentation demonstrated that his extensive ion optical calculations resulted in a much improved design of the vacuum chambers thereby significantly increasing the transmission through the separator. An extremely valuable feature for chemists will be the possibility to invert the polarity of the quadrupole magnets from a DQ_hQ_v to a DQ_vQ_h configuration. This results in a much smaller image size in the focal plane allowing the coupling of much faster chemical separator systems to TASCA. In his contribution, C.E. Düllmann [64] highlighted a number of new chemical systems that are well suited to be applied behind a pre-

separator. Of particular interest appears the future synthesis of volatile organometallic transactinide compounds i.e. Rf-hexafluoroacetylacetonate complexes, that cannot be accomplished without pre-separation. Another, potentially very interesting experiment requiring pre-separation was discussed by H. Hummrich. Elements $Z \geq 108$ may behave like noble metals and probably easy electrodeposition is possible. Short-lived ^{188}Pb nuclides were transported via a KCl-aerosol gas-jet to the collection system ALOHA where the collected activity was transferred quasi-continuously to solution. The solution was transferred to an electrolytic cell and Pb was electrodeposited on Ni(Pd) tape. Every 25 s the tape was stepped in front of 3 PIN diodes, where the α-particle decay of ^{188}Pb was registered. A contaminant that is produced in the synthesis of superheavy elements are Po isotopes that would electrodeposit very easily and give rise to a significant α-particle background. Thus, such an experiment would be run preferably with a pre-separator such as TASCA. An important contribution of chemists to the field of transactinide research is also the fabrication of suitable targets for both heavy ion irradiations as well as for laser spectroscopic investigations. In his poster contribution, K. Eberhardt presented two methods namely electrolytic deposition and moclecular plating that were applied to the preparation of exotic actinide targets.

4 Atomic and ionic level structure

The elements between thorium ($Z = 90$) and lawrencium ($Z = 103$) belong to the actinides, the chemical homologue elements of the lanthanides. These elements, in which the $5f$, $6d$, $7p$ and $7s$ electrons outside the closed radon core ($Z = 86$) form the electronic configurations, are of particular interest for the optical spectroscopy discussed in this workshop. As a rule, the $5f$ shell is filled in the actinides successively with increasing charge number of the nucleus. Hartree-Fock calculations show [65] that $5f$ electrons see a larger effective charge than $6d$ and $7p$ electrons. Consequently, most of the binding energy in the electronic configurations comes from the electrostatic attraction between the individual $5f$ electrons and the nucleus. This actinide contraction stabilizes the $5f$ configuration with increasing charge number. On the other hand, as already pointed out in Section 3.1, relativistic effects destabilize the f-electron binding and stabilize s and $p_{1/2}$ orbitals. These relativistic effects may change the atomic level structure in comparison to the homologue elements and is, as in chemistry, also one of the main research objectives in the transactinide atomic physics.

4.1 Relativistic effects and quantum electrodynamics

Relativistic effects and Quantum Electrodynamics (QED) gain importance for the heaviest elements. The resulting changes in the electron orbital energies become observable even for valence shell electrons. P. Pyykkö disentangled in

his review talk kinetic relativistic effects, the Breit interaction and the lowest-order QED contributions, i.e. the self-energy (SE) and the vacuum polarization (VP) which are the leading terms of the Lamb shift [66]. The VP can be derived from the Uehling potential [67], an attractive, local potential which is equal for all elements. The SE has the opposite sign and is larger than the VP. Kinetic relativistic effects on the valence shell behave roughly as Z^2, stabilize the ns shells due to their non-vanishing overlap with the nuclear core and destabilize the $(n-1)d$ shells. Lamb shift and the Breit contribution in the valence-shell are of the same order of magnitude, destabilize the ns-shells and indirectly stabilize the $(n-1)d$-shells, counteracting the kinetic relativistic effects at a $1-2\%$ level.

I. Goidenko presented investigations of the $7s$ Lamb shift of element 112 using the effective local-potential approximation as well as self-consistent approaches. It was found that the chemical reactivity of Eka-Hg is raised by these QED effects which are of the order of the $7s - 6d$ energy difference. The results are published within this volume [68]. For two-atomic molecules like Rg_2 the advantages of finite element calculations were demonstrated by O. Kullie [69].

P. Indelicato elaborated the difference of studying QED effects on inner shells and on the $7s$ valence orbits of the heaviest elements. While the quality of calculations of the $1s$ Lamb shift and K, L, and M shell ionization energies can be evaluated by the observation of characteristic X-rays [70], the corresponding effects on the valence shell become accessible by the experimental techniques discussed during the symposium. The resulting level shifts in the $7s$ shell of about 680 cm^{-1} in Rg should be easily detectable even though the ratio of the $7s$ SE to the $1s$ SE only amounts to 10^{-4}. However, while the SE of a point-like nucleus is the dominant contribution in the $1s$ shell, Welton screening [71] and the finite size of the nucleus can not be neglected considering the SE of the $7s$ shell. Studies of effects caused by the valence electrons therefore may help to determine contributions by relativity as well as by quantum electrodynamics.

4.2 Optical spectroscopy of atoms

It was the conviction of physicists and chemists in the fifties of the last century that it ought to be possible to deduce many of the properties of an element and its compounds from a detailed knowledge of its electronic configurations. The background behind this fact is that the trans-uranium elements Np, Pu, Am, Bk, Cm, Es, Fm, which belong to the group of the actinide elements, could at that time be bred only in relatively small amounts in nuclear reactors or were found in the fall-out of atomic bombs. Big programs were launched to investigate the atomic emission spectra. The spectra were studied with light from electrode-less quartz discharge tubes with samples of about 0.1 mg. As devices the 30 ft Paschen-Runge spectrograph at Argonne or the Fourier-transform spectrometer in the Laboratoire Aimé Cotton, Orsay, France, were employed. In each actinide spectrum tens of thousands of spectral lines can be observed. The levels are organized into terms, some dozen of terms form a configuration, and there are a dozen or more configurations. The order of hierarchy is not evident since there is considerable overlapping of different configurations and since the terms are not pure in any coupling scheme they must be described as mixtures (configuration mixing). In most cases the levels can be identified only by comparison with theoretical calculations. Corroborative evidence can be obtained by Hund's rule, isotope shift, or hyperfine splitting [65].

Data basis of atomic levels and transitions [72] represent the starting point for exploring the complex atomic structure of trans-uranium elements. It is also the backbone of many applications such as resonance ionization spectroscopy for the determination of ionization potentials of trans-uranium elements or for most-advanced ultra trace analysis of plutonium for various environmental tasks [73]. In his talk, J.V. Kratz explained how the element-selective resonance ionization method has been combined with mass-selective ion detection employing a time-of-flight mass spectrometer. A detection limit as low as 2×10^6 plutonium atoms of a single isotope has presently been reached which enables the identification of the origin of plutonium samples via a determination of its isotopic abundances.

From the spectroscopic measurements mentioned above quite good spectroscopic information is available up to the element einsteinium ($Z = 99$). For the element fermium ($Z = 100$) only the ground state was identified by Landé g_J-factor measurements to be a $5f^{12}\,7s^2\,{}^3H_6$ term [74]. No other information was available up to recently. The reason is obvious: on the one hand, the amount of material bred in nuclear reactors becomes less and less the more neutrons must be captured, i.e. the larger the atomic number gets. As a consequence, the conventional optical spectroscopy gets more and more difficult. On the other hand, elements with $Z > 100$ can be produced only in nuclear fusion reactions with very low rates. Novel ultra-sensitive experimental methods were required which were developed only recently. As a consequence, only lately the spectroscopic investigations launched in the fifties of the last century could be continued successfully.

Fusion product recoil separators like the SHIP facility at GSI in Darmstadt, in combination with traps in which fusion products can be stored, open a rich variety of new research opportunities in the heavy element atomic spectroscopy. The simplest conceivable trap is a buffer gas cell in which the fusion products come to rest either as atoms or singly charged ions in the surrounding of innert gas atoms like helium or argon. The capture time of the neutral atoms is determined by the diffusion process to the walls. This time is typically in the order of 50 ms and is therefore long enough to investigate unknown atomic level schemes or to determine the ionization potential of heavy elements by laser spectroscopy employing pulsed lasers with repetition rates of 200 Hz or more. Very powerful methods have been developed like the resonance ionization spectroscopy with detection of the ionization process either by radioactive decay [75] or directly

by the ions after mass analysis [26]. One of us (H.B.) gave an overview of these methods which were developed and tailored in the late eighties and early nineties for the measurement of the isomer shift of americium fission isomers. The very high sensitivity of this method enabled also the spectroscopy of elements for which no atomic level information was available [76]. K. Lauth and P. Kunz presented the status of a challenging laser spectroscopy experiment of the element nobelium ($Z = 102$) which is pursued in a collaboration of groups from the Universities of Mainz and Munich, and GSI. The method of resonance ionization spectroscopy with α-decay detection will be employed. Using a specifically designed buffer-gas cell filled with argon at SHIP, the collaboration could reach the required overall efficiency of about 1% in test experiments on the α-active chemical homolog ^{155}Yb [77].

Calculations for No and Lr have been performed in order to constrain spectral intervals to be searched for an optical resonance. These are based on Multi-Configuration Dirac-Fock calculations, as described in the talk of S. Fritzsche for the element lawrencium [78]. Accurate calculations of the excitation energies are a challenge for modern ab-initio theories. Apart from strong relativistic and quantum electrodynamical (QED) effects, most heavy elements must be treated as systems with several open shells. This applies particularly for those atoms and ions which involve open d and/or f shells. In addition, further difficulties arise from the large *number of electrons* which have all to be taken into account in any ab-initio theory [79]. These facts limit the accuracy for nobelium and lawrencium in favorable cases to less than ± 1200 cm^{-1} [78,80]. Nobelium has a closed [Rn] $5f^{14}7s^2$ ground state configuration which makes it an ideal candidate for the coupled cluster approach. U. Kaldor demonstrated the increase in the accuracy of Fock-space coupled cluster calculations [81] by the development of Intermediated Hamiltonian schemes [82]. For simple systems like alkalis and alkaline earth atoms, transition energies accurate to a few meV are obtained. A. Borschevsky presented her calculations concerning transition energies, ionization potentials and electron affinities of No and Lr. The expected uncertainty of the calculations of 0.09 eV $\hat{=}$ 700 cm^{-1} was deduced from analogous calculations for the lighter chemical homologues Yb and Lu for which the transition energies are known. A detailed description can be found within this volume for lawrencium [83] and for nobelium in reference [84]. The extension of this method towards other open-shell heavy and superheavy elements using extrapolated Intermediate Hamiltonians [85] was presented by E. Eliav.

The differences encountered between the properties of individual atoms in their atomic gaseous state and those in their condensed state were presented and discussed by R.G. Haire. In the condensed state, the $5d$- and $6d$-shells are energetically favored for bonding for most of the lanthanides and more than half of the actinides. But, in many instances, these d states must be made available through the promotion of an f electron. Consequently, the ground state of Cf ($Z = 98$) changes from $5f^{10}7s^2$ in its atomic

gaseous state to $5f^96d^17s^2$ in its condensed state after this promotion occurs. In contrast to the lighter actinide metals, the transplutonium elements do not have itinerant $5f$ electrons — their $5f$ electrons do not contribute significantly to their bonding or cohesive energies at atmospheric pressure — they are "softer" and display structures and phase transitions comparable to the lanthanides and transition metals. However, after delocalization of their $5f$ electrons by applying pressures of up to 100 GPa, the elements Am up to Cf acquire structures similar to the lighter actinides which now display less compressibility (e.g., have a larger bulk modulus). In essence this change may be thought of as a form of "modern alchemy": for example, the atomic volumes and physical properties of Am are transformed above 16 GPa into those observed first for gamma Pu and then alpha uranium [86,87].

4.3 Towards optical spectroscopy of ions

The SHITRAP facility allows fusion products, separated in the recoil separator SHIP from the primary beam, to be transferred by the use of linear Paul traps into a Penning trap system for nuclear mass measurements, see Section 2.3. Once singly charged ions are stored in Paul traps they may be illuminated by lasers. As shown in first experiments by P.E. Toschek, H. Dehmelt, W. Neuhauser et al. [88,89] and later by R.G. DeVoe and R.G. Brewer [90], the re-emitted fluorescence light is sufficiently intense to observe even single ions. However, such experiments may be difficult if no information on the ionic level structure is available. Even if a laser would be tuned on a resonance one has to cope with the rapid depopulation pumping into metastable low-lying levels resulting in a loss of the signal. A road map how to proceed in this kind of ion spectroscopy was presented by G. Werth and is published within this volume [91]. In particular he showed that metastable levels can be quenched by buffer gas collisions at a pressure in the order of 10^{-5} mbar. K. Wendt presented plans for a feasibility study of optical spectroscopy on the chemical homologue of nobelium, the element ytterbium, in a gas filled RFQ trap in which laser ionized species can be collected, cooled and bunched.

Model-independent information on properties of nuclear ground or isomeric states, as their spins, magnetic moments, quadrupole moments, and changes of the mean square charge radii, can be obtained from the hyperfine structure of optical transitions. P. Campbell presented the opportunities at the Cyclotron Laboratory of the University of Jyväskylä for laser spectroscopy on ions and neutral atoms for heavier element spectroscopy. A particular project at this facility aims to study nuclear isomerism in actinide elements and to provide pure accelerated beams of exotic nuclei by laser ionization in the source.

U. Schramm and M. Drewsen discussed the interaction of charged ions or molecules with Coulomb crystals of temperatures in the few mK range for the purpose of sympathetical cooling [92]. M. Drewsen presented a scheme for the detection of single-photon scattering event in a superheavy element using quantum logic of a system of two ions

in a linear Paul trap the motion of which are coupled by their Coulomb interaction. Such a scheme would enable a search of resonances in superheavy elements of sufficient long lifetime [93]. U. Schramm presented plans for a table-top laser based X-ray free electron laser [94] which enables access to optical features of heavy elements by excitation of inner shells.

5 Ion chemical reactions and ion mobility in buffer gases

One experimental difficulty is common for all "ion catcher" concepts based on buffer gas cells, namely that ions in the gas phase are chemically extremely reactive. Already smallest amounts of gas impurities may transfer the few ions in unwanted ion chemical compounds. Indeed, experiments at IGISOL-facilities have shown that ion signals often are fragmented over many masses. Yu. Kudryavtsev gave a summary of the extensive investigations performed at the Leuven Isotope Separator On-Line laser ion source, see e.g. references [95,96]. It was observed that the formation of ion chemical compounds with water is the dominant reaction mechanism even if gas impurities are reduced into the sub-ppb level. The binding energy of these hydrates lies in the eV region and it is possible to dissociate most of them in an acceleration field at a suitable buffer gas background pressure.

Buffer gas cells also provide the means for controlled ion chemical reaction studies in the ionic ground state [97, 98]. Controlled admixtures of gas impurities permit determining the reaction constants of ion chemical reactions like $Er^+ + O_2 \rightarrow ErO^+ + O$. This method appears extremely suitable for experiments at superheavy elements since principally only few ions are needed. On the one hand, ion chemical reactions interfere with subsequent experiments using atomic ions since their extraction efficiency is decreased. On the other hand, these observations may permit specific interaction studies of superheavy ions with noble gases and gas admixtures which contribute to a better understanding of the electronic structure in strong nuclear fields. A combined experimental and theoretical session was dedicated to the question what we can learn from ion chemistry of superheavy elements and how experiments could look like.

P. Indelicato discussed the combined effect of relativistic and QED effects on theoretical ionic radii. A detailed report can be found within this volume [99]. The possibility to directly probe these theoretically predicted radii by ion mobility measurements was controversially discussed by L.A. Viehland who gave a review on this field, see reference [100], and M. Laatiaoui [101].

The ion mobility K is connected to the drift velocity v_d of ions drifting in the in electric field E inside the gas cell via the relation $K = v_d/E$. Ion mobility measurements are a well established technique [102] for obtaining detailed information about interaction potentials of atomic ions in noble gases [103]. Recently, first experiments using trace amounts of short-lived actinides [97,104] have been

performed to study changes of the ion mobility for U^+ and Am^+ ions and for Cf^+ ($Z = 98$) and Fm^+ ($Z = 100$) ions after laser resonance ionization in an argon filled optical cell. The observed higher mobility of Fm^+ compared to Cf^+ was attributed to the smaller orbital size of the $7s$ orbital in Fm^+ due to the theoretically predicted actinide contraction. How this effect reflects in the mobility of mono atomic ions is discussed in this volume, see reference [101]. From the observed mobility of molecular ions detailed information about chemical binding such as bond lengths and contributing orbits can be deduced [105].

The connecting link between ab-initio calculations of a single ion and ion mobility are the interaction potentials between ions and the buffer gas atoms. While calculations for these potential have not been performed for the heaviest elements so far, respective calculations for lanthanides, the lighter chemical homologous of the actinides, have been carried through. A full discussion of the results, which have been presented by A.A. Buchachenko also for neutral lanthanides (LN), is given in this volume [106]. The striking result is that the interaction anisotropy in LN-He systems, arising from the the open $4f$ electronic shell, is largely suppressed by the closed outer shells [107]. This results in relaxation rates of populated Zeemann sublevels during buffer gas collisions which are small enough to allow magnetic trapping of LN atoms with nonzero angular momentum at background pressures of about 0.3 mbar of He. The result of these state-of the art ab-initio calculations is in accord with experimental observations reported in reference [108]. In this experiment it has been demonstrated that atoms can be cooled below 2 K within 5 ms after laser ablation by buffer gas cooling in ^3He at an initial He temperature of 350 mK. Similar behavior may be expected for actinides, so that the combination of buffer gas stopping cells with catchers, as described in Section 4 and reference [77], and subsequent laser ablation may pave the way for experiments at ultracold superheavy atoms.

A common challenging aspect in chemistry and ion chemistry are state selective reactions. P.B. Armentrout gave a review talk on this field and discussed how exited atomic and ionic states can be used to exert control over efficiency and specificity of chemical reactions [109]. By observing the onset of ion chemical reactions as function of the translational energy of the ions, detailed information about the electronic state of the ion can be deduced. Investigations at third-row transition metal ions, which are the chemical homologues of the transactinides, have shown that relativistic effects, including the lanthanide contraction of the valence s orbitals, lead to enhanced σ and π bonding with simple ligands. From these studies ion chemical reactions with oxygen and methane appear as promising systems for studies at actinides and transactinides.

Fundamental aspects of the chemical and physical properties of atomic and molecular actinide ions and neutrals are being examined by Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS). The experimental method as well as the wealth of experimental

data up to the element curium ($Z = 96$) has been outlined by J.K. Gibson on the workshop and is summarized in this volume [110]. The motivation for studying the properties of actinides in the gas phase lies in its direct relevance to theoretical studies [111,112]. Moreover, fundamental properties as e.g. $M^{+/2+}$-O and $OM^{+/2+}$-O bond energies can be deduced from metal ion oxidation reactions which help to illuminate the electronic structure of the heaviest elements. To continue experimental studies beyond Fm, the coupling of an FT-ICR detection scheme to a heavy isotope production facility is required as envisaged within the SHIPTRAP project at GSI.

6 Conclusion

Recent experiments with the gas-filled separators DGFRS (FNLR-JINR Dubna), GARIS (Riken, Tokai), RITU (University of Jyväskylä) and BGS (LNBL, Berkeley) as well as the velocity filter SHIP at GSI, Darmstadt have revealed a vast amount of information on the synthesis of superheavy elements and possibly discoveries of new elements up to the charge number $Z = 118$. Progress has been made as well in the nuclear spectroscopy of various fermium, mendelevium, nobelium, and lawrencium isotopes. In particular, the development of chemical separation techniques has permitted detailed studies of the nuclear structure around the closed deformed neutron shell $N = 162$ at hassium.

An outstanding experimental result was the chemical identification of element 112, confirming thereby the synthesis of superheavy elements in ^{48}Ca induced reactions. While it is still too early to draw final conclusions about the chemical properties of element 112, there are indications, that element 112 might be more volatile than its lighter homolog Hg. The success in identifying element 112 gives rise to the hope that also element 114 might be chemically identified in the near future. With the commissioning of the TASCA separator at GSI and similar setups at the BGS in Berkeley and at GARIS at RIKEN a new quality of chemical experiments with transactinide elements will become possible in aqueous- as well as in gas-phase.

Progress has been achieved in the optical spectroscopy of elements with unknown atomic level schemes. The recent development of very efficient laser spectroscopic techniques in connection with powerful atomic level calculations may pave the way for detailed precision studies of the atomic structure for the superheavy elements nobelium and lawrencium.

Buffer gas cell based concepts for stopping and extraction of nuclear fusion reaction products have been developed in recent years which will render first mass measurements of trans-fermium elements in ion traps possible in near future. Inherent to buffer gas techniques are interconnections with gas phase ion chemistry and ion mobility. Both techniques have already yielded detailed information about the electronic structure of the heaviest elements up to fermium which can be extended also to superheavy elements.

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