

The quest for superheavy elements and the limit of the periodic table

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ABSTRACT

The borders of the periodic table of the elements and of the chart of nuclides are not set in stone. The desire to explore the properties of atoms and their nuclei in a regime of very large numbers of electrons, protons, and neutrons has motivated new experimental facilities to create new elements and nuclides at the limits of atomic number and mass. But the small production rates and short lifetimes of superheavy nuclei and their atoms mean that ‘atom-at-a-time’ studies are the only experimental way to probe them. The physical and chemical data obtained so far, augmented by theoretical calculations, indicate significant deviations from extrapolations from lighter elements and isotopes. This situation raises the question: how much further can one push the limits of the periodic table? In this Review, we describe the major challenges in the field of the superheavies and speculate about future directions.

[H1] Introduction

Ever since Dmitri Mendeleev and Lothar Meyer introduced the periodic table of the elements (PTE) in 1869 (refs^{1–3}) and Glenn Seaborg, Edwin McMillan and others that followed synthesized the first elements beyond the naturally-occurring uranium⁴, human curiosity has pushed the limit of the PTE to study the physical and chemical behavior of heavier and heavier elements. With the discovery of oganesson (atomic number $Z = 118$), the 7th row of the PTE is now complete⁵. However, the quest to reveal the region of superheavy nuclei exhibiting enhanced stability (the Island of Enhanced Stability, see Box 1) continues. What surprises are yet to be uncovered with a few extremely short-lived superheavy atoms produced at nuclear facilities at a rate of one atom every few months?

Studies of superheavy elements (SHE) with $Z \geq 104$ (Ref.^{6,7}) offer detailed insights into the nuclear and electronic structure in the presence of enormous electrostatic forces. The extent of the superheavy region towards higher atomic (Z) and neutron (N) numbers is unknown at present. From a nuclear physics point of view, exploring the unknown region of superheavy nuclei (loosely defined as atomic nuclei with proton number $Z \geq 104$) would help develop a deeper understanding of the nuclear landscape limited by the *nucleon driplines*, α -decay, and nuclear fission. The main difficulty in exploring the unknown regions of superheavy nuclei is to describe the competition between the strong force that produces nuclear binding and the electrostatic repulsion. The resulting Coulomb frustration at high nuclear charges is expected to give rise to exotic nucleonic topologies⁸ (that is, unusual nucleonic densities in the form of bubbles or tori) and impact both nuclear binding and decay modes and lifetimes of superheavy nuclei. From a chemistry point of view, the question is how these SHE behave chemically, if, and how, they fit into periodic trends, and how far the PTE extends⁹. Theoretically, precise calculations require the accurate treatment of *relativistic*, *quantum electrodynamics* (QED), and *electron correlation* effects. From the astrophysics point of view, short-lived

superheavy nuclei could be synthesized in the dynamical ejecta of neutron star mergers that are expected to be sites of the astrophysical *rapid neutron capture process* (*r*-process)^{10,11}. Such formed superheavy nuclei are expected to impact *r*-process abundances by recycling the nuclear material down to lighter mass regions via fission processes, thus offering a hint of SHE synthesis in the Cosmos.

What is the heaviest bound nucleus and the heaviest bound atom and what are their properties remain open questions. This Review primarily focuses on the recent developments in the field of SHE research. More in-depth discussions and references can be found in Refs.^{3,12–15}.

[H1] Synthesis and detection

The heaviest known elements (in fact all nuclei with $Z \geq 114$) have been produced in *heavy-ion induced fusion reactions* using beams of doubly-magic ^{48}Ca ions. In these reactions, nuclei of the ion beam have been fused with target nuclei of the appropriate actinide element to reach proton numbers $Z = 114 - 118$. Unfortunately, to go to higher atomic numbers, the ^{48}Ca strategy does not work due to the technical inability to produce sufficient amounts, on the order of 10 mg to 20 mg, of trans-Cf targets¹⁶. To reach $Z \geq 119$, ion beams heavier than ^{48}Ca are therefore needed. Different nuclear reactions were tried in months- to year-long searches for the elements $Z = 119$ and $Z = 120$, albeit discovery was not reported to date^{17,18}, with experiments still running^{19,20} (see also Refs.^{21,22} for planned campaigns).

A key challenge — for experiment and theory — is thus the identification of the most promising nuclear reactions and the optimum ion beam energy, which is a critical parameter. In addition, a successful identification of a superheavy nucleus requires that synthesized nuclei first, live for at least $\sim 1 \mu\text{s}$, which is the flight time from the point of creation and through the *recoil separators* (needed for isolation of the single atoms) into the detection system; second, decay by a mode that can be registered with high efficiency, such as α -decay or spontaneous fission (SF); and third, decay in a way that allows for unambiguous identification of the superheavy nucleus. This latter condition is much easier to fulfill in the case of α -decay than in SF, as the atomic numbers of the fission products still cannot be measured directly for a single decaying nucleus.

Alternative pathways have been suggested for the production of superheavy nuclei, particularly those that are more neutron-rich than can be achieved through actinide-based fusion reactions with stable-isotope ion beams. Indeed, as shown in Box 1, the predicted longer-lived superheavy nuclei are on the neutron rich side. The direct approach to the anticipated $N=184$ shell closure will only be possible at higher Z , using, for example, the $^{251}\text{Cf}(^{58}\text{Fe},n)$ reaction leading to $^{308}124$ (ref.²³). Alternatively, collisions of two massive nuclei, ^{238}U and ^{248}Cm , which lead to the exchange of a large number of nucleons between the projectile and target nuclei, have been experimentally explored²⁴ and ref.²⁵ suggested further studies. The idea of such *multinucleon transfer reactions* was picked up in exploratory studies²⁶.

Another avenue, which would in principle reach nuclei located more closely towards the center of the ‘Island of enhanced stability’ (see Box 1), expected to reside in the neutron-rich area of the chart of nuclides, involves accelerated *radioactive-ion beams* (RIB) of neutron-rich radioactive nuclei. In the early Ref.²⁷, the optimum pathway into this region was evaluated. In light of the new RIB facilities having come online in the past years (Radioactive Ion Beam Factory (RIBF) in Japan, Facility for Rare Isotope Beams (FRIB) in US) or being under construction (Facility for Antiproton and Ion Research (FAIR) in Germany, Rare isotope Accelerator complex for ON-line experiments (RAON), South Korea, Heavy-Ion Accelerator Facility (HIAF) in China, GANIL/Spiral-2 in France), an update using current experimental rates may help identifying the most exciting possibilities. Closer access to the region of highest stability might also be gained if electron-capture or β^+ -decaying nuclei of heavier elements were discovered^{18,28}. In this case, further guidance from nuclear theory is required to identify promising candidate nuclei, and experimental technical advances are needed to register such nuclei, as current setups are optimized to register α -decay and/or SF but not electron-capture or β^+ -decay. Once lifetimes become longer, decay-based identification becomes less efficient, prompting the need for single-ion registration capabilities in particle-traps, which can detect image currents induced by individual live ions²⁹. The nuclei produced in ‘hot fusion reactions’ with ^{48}Ca ion beams, that is all nuclei with $Z \geq 114$ and their decay products, are currently not linked to the known region of the nuclear chart because their α -decay chains terminate by spontaneous fission, as can be seen in the Box 1 figure. An important goal is to connect these nuclei to the nuclear mainland thus providing a direct mass/charge identification^{30,31}. Research to establish this connection is ongoing³².

Cross-sections of yet-undiscovered nuclei are generally expected to be very small. Consequently, experiments need to use more intense ion beams, thicker targets³³, and/or longer run-times. Upgrades of existing accelerators²⁰ and construction of new ones^{21,22,34} require advances in target production to accept the ever more intense ion beams over ever longer run-times. As the nuclear reaction pathway based on stable doubly-magic ^{208}Pb and neighbouring nuclei appears largely exhausted with only 3 atoms of ^{113}Nh registered over a total run-time of 553 days within 9 years³⁵, quantities of transuranium isotopes in the range of tens of milligrams are prerequisite for the further exploration of the superheavy region¹⁶.

[H1] Structure and stability

Once superheavy nuclei are created, experiments focus on the study of nuclear structure via the detection of prompt radiation emitted in the decay of excited fusion products and in the radioactive decay of superheavy nuclei into their ground or metastable nuclear states³⁶. Such studies inform about the nuclear stability landscape and the underlying nuclear shell structure. The evolution of binding energies is studied by high-precision mass spectrometry³⁷. Nuclear size and shape are probed by laser spectroscopy studies³⁸ albeit currently limited to the region around ${}_{102}\text{No}$, accessible in high cross-section reactions (in the order of $\approx \mu\text{b}$) of doubly-magic ${}^{48}\text{Ca}$ with doubly-magic ${}^{208}\text{Pb}$ and neighboring nuclei.

The known superheavy nuclei are all radioactive with α -decay and SF being their main decay modes. β -decay is a fundamental nuclear decay as it determines the stability of the majority of isotopes in the nuclear chart. However, since it is governed by the electroweak force, the corresponding half-lives are much longer than those due to α -decay or SF^{39–42}. Still, β -decay channels should not be ignored, especially for the proton-rich superheavy nuclei as synthesized to date⁴³. It is important to note, however, that according to a recent global analysis⁴⁴, forbidden β -decays, not considered in theoretical studies of Refs.^{39–42}, are expected to be very important in heavy nuclei. This calls for a revision of the theoretical estimates of β^+ /electron capture rates in superheavy systems.

As discussed in Box 1, according to current models based on realistic nuclear forces, no ‘stable superheavy island’ is expected theoretically. Since the superheavy nuclei inhabit the virtually-unexplored region of the nuclear chart, theoretical predictions amount to extrapolations. To this end, the predictive power of nuclear models in the unknown domain of complex superheavy nuclei, consisting of many hundreds of nucleons, can be improved by using high-performance computing and modern techniques and algorithms such as machine learning. One possible extrapolation strategy is to develop emulators based on training sets of deviations between experimental data and calculated observables (residuals). Such emulators can be constructed by, using *Bayesian machine learning*^{45,46} for example. Unlike the raw predictions of global models that vary widely, the quantified model predictions corrected by residuals obtained by means of statistical machine learning are very consistent⁴⁵. Such machine learning assisted extrapolations can be obtained for entirely unknown observables, such as masses or fission lifetimes. By adding new measurements to the training dataset, the fidelity of short- and medium-range extrapolations will be continuously improved.

When making extrapolations into the superheavy territory, however, one should be very careful when trusting extrapolative predictions of any given model because of systematic model uncertainties. By considering the collective wisdom of several models based on different theoretical assumptions and different calibration strategies, together with the most recent data on superheavy systems, one can apply powerful Bayesian model mixing technique to assess model-related uncertainties⁴⁷.

Figure 1a shows the posterior probability of existence for nuclei in the chart of nuclides based on predictions of eleven global mass models, the most recent data on nuclear existence and masses, and three model-averaging strategies⁴⁸. According to this analysis, limited to $Z \leq 119$, there should be between 6500 and 8500 particle-bound nuclei in this range of atomic numbers. The results of the individual models show considerable spread around the neutron dripline due to the extrapolation uncertainty in the heavy neutron-rich region. For more examples of current global nuclear calculations aided by machine learning see Ref.⁴⁹.

Extrapolations beyond $Z = 118$ are going to be difficult because of an increased competition between the short-ranged nuclear force and the long-ranged electrostatic force. The associated Coulomb frustration effects are expected to result in exotic topologies of nucleonic densities, such as bubbles, rings, or tori^{8,50,51}. An example of anticipated exotic forms of nucleonic matter in the superheavy region is shown in Fig. 1b for the superheavy nucleus with $Z = 156$, $A = 466$. The spherical local minimum (marked ‘D’) has bubble-like density distribution with a pronounced central depression. The lower-energy configurations ‘A’ and ‘B’ correspond to toroidal forms. At the local minimum ‘C’ the nucleus ${}^{466}156$ has a very flat oblate shape while it is very elongated (prolate) at ‘E’, and ‘F’. The stability of the local minima, having exotic density distributions to triaxial and reflection-asymmetric distortions, is yet to be investigated comprehensively^{50,52,53} as many-dimensional collective spaces need to be explored. Due to this difficulty, the extent of the superheavy region is still unknown. Again, high-performance computing and machine learning will be essential for tackling this question.

Apart from the difficulties related to the frustration effects, there is a considerable model-dependence when it comes to SF modeling. This point is discussed in Box 1 and also illustrated in Fig. 1c, which shows the fragment charge distribution predicted for four recent theoretical models of Refs.^{54–56} for the known superheavy nucleus ${}^{294}_{118}\text{Og}$. Indeed, a quite dramatic model dependence of the predictions of SF yields is seen. The model of Ref.⁵⁴ predicts highly asymmetric fission, or cluster emission. However, other models^{55,56} predict fairly symmetric fission-yield distribution. This discrepancy calls for an experimental search for a cluster emission from ${}^{294}\text{Og}$ and other superheavy nuclei.

At the final stages of the astrophysical r-process, neutron-rich heavy and superheavy nuclei could impact the formation of elements heavier than iron through the fission recycling – believed to be of particular importance during neutron star mergers where high free neutron number densities are available^{10,11,57,58}. Yet, due to considerable model uncertainties pertaining to the modeling of fission and β^- -decay rates, there is no consensus regarding the role of the superheavy nuclei in r-process

nucleosynthesis^{10,11}.

[H1] Electronic structure and location in the PTE

Atomic physics experiments with single atoms rely heavily on theoretical predictions from atomic structure calculations. There are currently many obstacles on the road to the required accuracy to evaluate and compare the total binding energies of different atomic states to find, for example, the correct electronic ground state. An added challenge is to predict the transition energies well enough to allow for finding these transitions more easily in experiments³⁸.

Up to now, laser spectroscopy experiments were successfully performed for the elements up to $_{102}\text{No}$ (Ref.⁵⁹), and work is in progress⁶⁰ for $_{103}\text{Lr}$. However, laser spectroscopy advances³⁸ beyond $_{102}\text{No}$ are currently limited by the large uncertainties of atomic physics predictions of the electronic shell structure, which hamper the development of laser excitation schemes for resonant ionization of the isotopes of heavier elements. Progress in the prediction of the hyperfine structure and isotope shifts is also necessary. Calculations are now available for einsteinium⁶¹, fermium⁶² and nobelium⁶³. This is a prerequisite for detailed studies of their atomic structure and of nuclear structure via hyperfine structure studies. Such information on the atomic level structure may ultimately aid in the optical identification of such heavy elements as the products of astrophysical r-process events⁶⁴.

The method of Resonance Ionization Spectroscopy (RIS)^{65,66} was adapted for studies of single atoms produced in fusion-evaporation reactions where detection is achieved by registering the radioactive decay of an ion produced by resonant laser-ionization³⁸. A first breakthrough was achieved⁵⁹ in 2016 in $_{102}\text{No}$ with the identification of a suitable excited state for two-step photoionization after several years of scanning across the theoretically-predicted wavelength range. This laid the basis for precision studies of the nuclear⁶⁷ and atomic⁶⁸ structure of nobelium isotopes. Technical advances focus on obtaining higher-precision data by implementing improved techniques^{69,70} and on expanding the range of accessible isotopes to species with lifetimes outside the bounds of the initial incarnation of the technique^{60,70,71}. One of the key problems in advancing laser spectroscopy studies to heavier elements is the need to search for electronic excited states that allow efficient resonant ionization online using rare isotopes produced at minuscule rates. To aid the experimental identification of an atomic level suitable for resonance ionization in an atom where no single such level has been known, theoretical guidance is of utmost importance. Unfortunately, theoretical uncertainties exceed typical laser bandwidth substantially, leading to very long scanning times in the search of the location of the level for each and every new element that shall be probed by RIS. An example for the laser spectroscopy of $_{102}\text{No}$ is discussed in Ref.⁷². There is a large dispersion $\approx 0.15\text{ eV}$ on the transition energy of $\approx 3.5\text{ eV}$ in theoretical transition energy evaluations, depending on the method used to evaluate electron correlation including core-valence interactions, as well as QED and nuclear size effects⁷³⁻⁷⁶. Predictions exist for heavier elements (Lr, Og)^{74,77}, but are bound to be even less accurate.

Because of the very large *relativistic effects* encountered in SHEs⁷⁸, electronic structure calculations must rely on the Dirac relativistic formalism including bound-state quantum electrodynamics (BSQED)⁷⁹. The structure of the chemically-relevant outer atomic shells critically depends on relativistic effects, even for lighter elements where, for example, a delicate interplay between the $(n-1)d$ and ns shells takes place in the transition metals⁸⁰. The ratio of relativistic and BSQED corrections to nonrelativistic energies in atomic or molecular properties scales approximately as $(Z\alpha)^2$, where α is the fine-structure constant. Moreover, orbital mean radii change very differently depending on Z and on the angular momentum, thus modifying their interaction with other electrons (see for example Ref.^{81,82}) leading to significant changes in their electronic energies. Hence, in the case of the SHEs, these effects strongly influence the electronic structure, making it very difficult to determine their correct position in the PTE. Large deviations from properties derived from extrapolations within a group of the PTE are therefore expected.

For finding the correct ground state and transition energies from excited electronic states, one must reliably evaluate the electron correlation energy using the electron-electron interaction operator derived from BSQED. Correlation effects must then be included, by using relativistic many-body techniques, such as the Multiconfiguration Dirac-Fock method, the relativistic configuration interaction method or relativistic many-body perturbation theory⁸³. Moreover, one should also take into account BSQED contributions of order $(\alpha/\pi)(Z\alpha)^4$ (self-energy– SE and vacuum polarization– VP), and of order $(\alpha/\pi)^2(Z\alpha)^4$ (two-loop SE, mixed VP-SE, multi-loop VP)⁸⁴. Many of these contributions have never been calculated for the high- n and high- j shells that are occupied in the SHE. Although the VP, at least at the Uehling, Wichmann and Kroll and Källén and Sabry level of theory^{84,85}, can be incorporated in calculations to obtain mixed-VP-multi-electron effects, the SE is not naturally amenable to such calculations. Direct BSQED calculations have been performed only for $n \leq 2$ orbitals⁸⁶, and model operators were developed⁸⁷⁻⁹⁰ allowing for an approximate incorporation of the SE. Such operators, including finite nuclear size corrections, have been applied⁹⁰ for the elements up to $Z = 170$ improving the precision of electronic structure calculations. Yet, it is not known how large the missing QED contributions are when using such model operators. The largest error in the predicted atomic spectra, however, most likely comes from the insufficient treatment of electron correlation.

There are several unresolved issues that make the electronic structure problem even more complex. When performing

numerical calculations, using the Multi-Configuration Dirac-Fock method, one must keep the occupied orbitals of the same symmetry orthogonal. Yet, when calculating a specific level of a given total angular momentum J for a very high Z , the electronic configuration tends to correspond to an almost pure jj coupling. For example, for a $nsn'p_j$ ($J = 1$) level, the lower eigenvalue (3P_1 at low- Z) is mostly $nsn'p_{1/2}$ ($J = 1$), whereas the upper is mostly $nsn'p_{3/2}$ ($J = 1$). The mixing coefficients are then close to 1 and 10^{-4} respectively⁷⁴. One of the two $n'p_j$ orbitals behaves like a correlation orbital, and one has to relax the node-counting constraint on it. This is true for any configuration with open shells. The definition of a bound versus a correlation orbital becomes blurred, and this can lead to severe convergence problems even for cases without electron correlation.

The difficulty of extending correlation calculations to the SHE also lies in the dense energy spectrum. To obtain electron correlation contributions, one has to perform single, double, and, eventually, triple and quadruple excitations to the unoccupied orbitals in the configuration interaction (CI) procedure. The latter two correspond to higher-order corrections and may be required to achieve better accuracy for the spectrum. Proper treatment of single excitations may be required to get the correct fine-structure values and non-relativistic limits⁹¹. However, for the SHE both occupied and unoccupied shells can have rather large j values ($j = 7/2$ for f shells, $j = 9/2$ for g shells), leading to huge numbers of quasi-degenerate configurations and thus very large Hamiltonian matrices and even larger numbers of Coulomb interaction coefficients between configurations. Even worse, when taking into account the Breit interaction⁹², which accounts for magnetic and retardation effects between two electrons, the number of interactions coefficients can be 2 orders of magnitude larger. Therefore, new methods and algorithms need to be developed to select configurations with appreciable contributions.

Recently, an extensive relativistic study of the ground states of the SHE in the range $120 \leq Z \leq 170$ was performed⁹³. In this work, the Breit interaction was taken into account and QED effects were considered within a model operator approach. It was found that the Breit interaction can change the ground-state configuration, whereas the SE and VP effects do not. It was also found that, in the case of energetically close configurations, electron-correlation effects can change the dominant configuration of the ground-state level. The deviations in predicted ground state configurations between the single relativistic configuration (SRC) and the CI approach are mainly concentrated in the range $Z = 131 - 138$, where the $5g_{7/2}$ and $5g_{9/2}$ shells are partially occupied and strong interactions between several configurations take place. A more accurate treatment of the electron correlation resulted in changes of the ground-state level structure in 4 of the 51 SHE considered. The main contribution to the ground state configurations of the SHE by means of the CI method is shown in Fig. 2. The PTE that can be drawn from this is shown in Fig. 3 for the heavier elements. This PTE is rather different from the one in Ref.⁹⁴ done at the Dirac-Fock level and where, for example, the $6f$ row goes from $Z = 141$ to $Z = 155$ and the $5g$ row is from $Z = 121$ to $Z = 138$. For $Z = 168$, Ref.⁹³ showed that the variation of energy due to different methods used to evaluate electron correlation can change the ground state level between $9s_{1/2}^2 8p_{3/2} 9p_{1/2}$ ($J = 1$) and $9s_{1/2} 8p_{3/2}^2 9p_{1/2}$ ($J = 1$). Using the *MDFGME* multi-configuration Dirac-Hartree-Fock code⁹⁵, and adding all single, double and triple excitations from $8s^2 8p^4 9s^2$ to $9p$, $8d$, $7f$ orbitals, with the Breit interaction treated fully self-consistently, leads to a $9s_{1/2}^2 8p_{1/2}^2 9p_{1/2}$ ($J = 0$) ground state configuration. The same ground state configuration is obtained by using the Breit operator and Uehling potential perturbatively⁹⁵. However, using the Breit operator self-consistently and the Uehling term as a perturbation leads to a $9s_{1/2}^2 8p_{1/2}^2 9p_{1/2} 8p_{3/2}$ ($J = 1$) ground state. Yet, the energy difference between the different possible ground states is quite small compared to the correlation energy, severely limiting the possibility to predict the correct ground state. Improvements in electron-correlation methods to better understand which contribution impacts the energy levels is therefore highly desirable.

Electronic structure calculations are limited to $Z \approx 171$, as beyond this nuclear charge the radial Dirac equation predicts that the $1s$ shell merges with the *negative energy continuum*. It has been argued that the resonance of an empty orbital with the continuum could lead to the spontaneous decay of the vacuum into a positron and an electron⁹⁶. The observation and measurement of such a process would lead to a much better understanding of the QED vacuum and of strong-field physics, including pair production in black holes⁹⁷. This critical regime can be reached for example by the collision of two uranium nuclei^{98,99}. Theoretical calculations have been carried out in order to distinguish the spontaneous pair creation from dynamical pair creation^{100,101}, the latter of which is induced by a strong time-dependent electric field created by the colliding nuclei. Whether this implies the end of the PTE from an electronic point of view is still being debated⁹. In our opinion, a filled $1s$ shell embedded in the negative energy continuum must be described within a rigged Hilbert space formalism, and from an electronic point of view there is no end to the PTE. It is hoped for that a quantum theory for multi-electron systems can be successfully developed for such exotic diving states. See Ref.⁹ for more detailed discussions.

[H1] Atom-at-a-time chemistry

Understanding the properties of SHE¹⁰²⁻¹⁰⁴ and the structure of the PTE motivates chemical studies¹², which have been performed for all elements up to $_{108}\text{Hs}$. Leaving a gap at $_{109}\text{Mt} - _{111}\text{Rg}$, these have recently reached $_{112}\text{Cn}$ to $_{114}\text{Fl}$; for the latter, the isotopes $^{287-289}\text{Fl}$ were used, having half-lives in the range of one second and being available at production rates of few atoms per day. More generally, all studies of the accelerator-produced elements face very low production rates and short

nuclear lifetimes; this implies experiments with single atoms¹⁰⁵. As cross sections and half-lives in the SHE region generally decrease with increasing nuclear charge, the early transactinides (that is the elements beyond the actinides) such as $_{104}\text{Rf}$ have been studied more extensively in the gaseous and liquid phase. In the liquid phase, where complex formation takes place, these studies gained access to compound stabilities, hydrolysis behavior and ionic radii¹⁰⁶. Conclusions have typically been drawn from the comparison of data obtained for the heaviest elements against similar data for their lighter homologs, also using short-lived radioisotopes in sufficiently small quantities to be in the regime where single atoms cannot interact with others of their kind. To obtain most impactful comparative data, one has to ensure that studies of all homologs are carried out under identical conditions.

Recent experiments have advanced to ever heavier elements, accessible at minuscule production rates and having half-lives of around a second. The lightest transactinides, though, can be produced at comparatively high rates, and isotopes with longer half-lives of typically tens of seconds are known. Therefore, much more detailed studies are possible that give access to a wider set of chemical systems and observables. The detection of single atoms always relies on the unambiguous detection of nuclear decay — most often α -decay chains. In liquid-phase studies, the liquid has to be evaporated before the α -measurement can be performed. This rather time-consuming step has so far prevented studies of elements beyond $_{106}\text{Sg}$ but inspired technical advances^{107,108}, which open up new avenues for future liquid-phase studies. In the gas-phase, historically the focus has been on simple volatile compounds such as the SHE halides in which the SHE is in its highest oxidation state.^{12,109} Technical advances allowed for the synthesis of a new compound class, a Sg carbonyl compound, featuring a metal-carbon bond typical of an organometallic compound^{110,111}. Work towards the characterization of the first bond-dissociation energy has started¹¹², holding the promise to give new insight into the influence of relativistic effects on the chemical behavior of this SHE. Further work has shown that mononuclear carbonyl complexes also exist in the groups 7–9 of the PTE, thus opening up an avenue for the chemical characterization of the yet uncharacterized $_{109}\text{Mt}$ (ref.¹¹³).

Atom-at-a-time chemistry experiments rely heavily on theoretical predictions from atomic and molecular structure calculations. However, in contrast to atomic structure calculations, wave function theory based methods for large molecules or infinite systems such as the solid state or for (chromatographic) adsorption studies on surfaces become computationally infeasible and one has to resort to less accurate relativistic density functional theory (DFT)^{114,115} or semi-empirical methods instead¹². As there is a variety of density functional approximations around¹¹⁶, it is advisable to test their performance against either accurate relativistic ab-initio calculations or available experimental data for the lighter group members^{117–120}. Moreover, in SHE adsorption studies on specific surfaces such as gold or quartz, London-type dispersion interactions need to be included¹²¹, for example through the Grimme's dispersion corrected DFT¹²². For applications on dispersion corrected relativistic DFT to SHE adsorption studies see Refs.^{121,123,124}. A future computational challenge will be to accurately model the adsorption dynamics on realistic surfaces, as present in the experiment, to obtain reliable thermodynamic data. The surfaces in such experiments are often not ideal surfaces such as Au(111) considered in theory work. For example, an analysis of the Au surface present in adsorption studies of Fl revealed this to consist mostly of polycrystalline Au grains and some well-ordered epitaxial Au(111) crystallites¹²⁵.

Advances in the region of the heaviest elements depend on automated chemistry systems that provide the highest efficiency for studies with isotopes that live for less than a second. One successful avenue is based on physical preseparation¹²⁶ of nuclear fusion products from unwanted byproducts of the nuclear reaction that would interfere with the unambiguous detection of a single atom. Preseparated single atoms are thermalized in gas, which flushes them into gas-chromatography systems, in which the chromatography column consists of silicon detectors suitable for registering the nuclear decay. The detectors are covered with thin layers of the material of choice on which the chromatographic process takes place, that is, mostly gold and silicon oxide¹²⁷, with selenium being explored as a further option¹²⁸. This approach¹² has allowed studies also of $_{114}\text{Fl}$. Probing the interaction strength with a gold surface has led to some controversy, whether Fl behaves like a rare gas¹²⁹ or like a metal¹³⁰; new data point towards a more metallic character¹²⁵, in contrast to earlier predictions¹³¹, but in line with theoretical work^{120,121,132}. However, one should be careful here as, very much like Hg (ref.¹³³), the bonding between Fl atoms is expected to be of Van-der-Waals type, but in the solid state it should be a metal or a semiconductor with a very small band gap¹²⁰. These experimental methods in principle promise to allow studying $_{113}\text{Nh}$ and potentially $_{115}\text{Mc}$, but will then reach their limitations as the half-lives of the longest-lived known $_{116}\text{Lv}$ isotopes are in the tens of milliseconds range only. The most time-consuming step of current setups is the thermalization and flush-out process of the thermalization chamber. This proceeds much faster if electric fields are applied that drive the ions towards the chromatography channel¹³⁴, with current exploratory setups achieving extraction times of around 55 ms¹³⁵. Highly-optimized systems¹³⁶ and the advances in other techniques such as vacuum-chromatography¹³⁷ promise to reduce this time further. Once these developments come to fruition, chemical studies of $_{116}\text{Lv}$, produced in the $^{48}\text{Ca}+^{248}\text{Cm}$ reaction^{138–140} and $_{117}\text{Ts}$, produced in the $^{48}\text{Ca}+^{249}\text{Bk}$ reaction^{141,142} will be within reach.

Besides ever-shorter half-lives, further challenges need to be addressed to achieve progress in this direction. Current chromatography setups operate at column temperatures of at most 35 °C, dictated by the maximum operation temperature of

silicon-based detectors for nuclear radiation. The development of high-temperature compatible detectors, based on materials such as electronic grade single-crystal chemical vapor-deposited (escCVD) diamond detectors¹⁴³ will enable more detailed studies of more reactive heavier *p*-block elements. Although the elemental state is, by far, the most likely speciation of the rather inert elements $_{112}\text{Cn}$ (ref.¹⁴⁴) and $_{114}\text{Fl}$ (refs^{120,125,129}), as supported by theoretical calculations¹⁴⁵, this is different for the other *p*-block elements. In the case of $_{113}\text{Nh}$, beside the elemental state, the hydroxide molecule has been suggested to be present in past experiments^{146–149}, and has to be considered in the heavier elements $_{115}\text{Mc}$, $_{116}\text{Lv}$, and $_{117}\text{Ts}$ as well.¹⁵⁰ Hence, fixing the speciation will be important in future studies of these elements. Naively, one could expect that the next member of the noble gas group, $_{118}\text{Og}$, would behave in a similar way to Rn. This element has become the focus of intense theoretical work^{117,119,151–153}. In line with the increasing chemical reactivity down this group, but in contrast to the notion of Og being a true noble gas, theory suggests that Og exhibits a positive electron affinity^{153–155} and is neither noble nor a gas¹¹⁹. It should also be more reactive towards Au compared to Rn, and perhaps also forming OgOH (refs^{150,156}). This might be probed in experiments similar to those already performed¹²⁵ for Fl. However, only a single isotope of this element has been detected so far, ^{294}Og with a half-life of about 1 ms (ref.¹⁵⁷), which is at present outside of the reach of even the most advanced currently conceivable chemistry experiments. More long-lived Og isotopes are expected to exist, with the half-lives increasing with increasing neutron numbers, in accordance with the experimental trend observed in isotopic chains around similar neutron numbers¹⁵⁸ in $_{110}\text{Ds}$ to $_{117}\text{Ts}$ (see inset in the Box 1 figure for the trend in even-*Z* elements). More neutron-rich, and potentially more long-lived, Og isotopes are expected to be accessible in reactions with more neutron-rich Cf target nuclei including ^{250}Cf and ^{251}Cf . Moderate samples (mg) of these isotopes are available in an isotopic mixture with ^{249}Cf as remnants of legacy ^{252}Cf spontaneous fission sources, in which the comparatively short-lived ^{252}Cf has decayed. However, a first search experiment using such a mixed-isotope target only observed ^{294}Og , likely from the ^{249}Cf component in the target¹⁵⁹. More sensitive studies become feasible once higher ion beam intensities¹⁶⁰ will be routinely delivered by the next-generation facilities^{20–22,34} as they become operational. Establishing actinide mass separation capabilities suited for providing isotopically pure targets of Cf, the heaviest element available in sufficient quantities, would substantially boost such efforts.

Can we push atom-at-a-time chemistry even beyond $_{118}\text{Og}$? It is hoped that the elements up to at least $Z = 126$ can be produced in the future and some may become accessible to chemical experiments. In the meantime, theoretical studies¹⁶¹ are the only way to predict the physical and chemical properties for the elements beyond Og. It is evident that such calculations will be challenging from a computational point of view as QED and electron correlation effects have to be properly accounted for, in atoms and in molecules. If successful, one can make more accurate predictions of where to place the SHE into the PTE.

[H1] Outlook

Our understanding of superheavy atoms and their nuclei has substantially improved in the last three decades, thanks to the discovery and study of elements with $Z > 109$. Laser spectroscopy and the determination of ionization energies for elements with $Z \geq 100$ has also provided valuable knowledge and tests of theoretical methods. However, even the most advanced use of theoretical methods in nuclear physics, with the help of machine learning techniques to deduce the properties of unknown nuclei from properties of known ones, have not yet enabled us to develop reliable theoretical frameworks to predict optimal experimental conditions for the synthesis of new elements and isotopes. On the atomic side, the prediction of the position of some elements in the PTE remains a major challenge. The precise calculation of transition energies on known SHE is extremely challenging, but is required to enable experiments to find these transitions and measure them accurately. But, as discussed in this Review, the experimental determination of the chemical properties of SHE has made enormous progress. Thus the study of the PTE, more than 150 years after it was first proposed, remains a vivid and exciting field of research that will generate new ideas and methods that will impact nuclear and atomic physics research, and chemistry.

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Key points:

- Experiments to synthesize new superheavy nuclei and elements beyond the heaviest currently known element oganesson are underway. These systems will be crucial for benchmarking and testing many-body atomic and nuclear theory.
- Rapid and efficient chemistry experiments with single atoms and molecules elucidate the influence of the high atomic charge on chemical properties, thus probing the fundamental architecture of the periodic table.
- The field of superheavy element research puts atomic and nuclear theory to the test. For many superheavy systems, all available information must come from theoretical extrapolations based on models aided by high-performance computing and machine learning.
- The presence of large electrostatic forces gives rise to pronounced relativistic effects in the atomic system and strong Coulomb frustration effects in the nuclear system. There are theoretical suggestions indicating that superheavy atoms should differ fundamentally from lighter species, leading to deviations from the current patterns of the periodic table.
- Fundamental difficulties are encountered when dealing with the many-particle Dirac equation, as beyond a certain nuclear charge levels such as the $1s$ are predicted to merge with the negative energy continuum, eventually leading to a potentially unstable atomic structure and real electron-positron pairs creation.

Box 1: An island of enhanced stability?

In the early days of nuclear physics, nuclei with 2, 8, 20, 28, 50, 82, and 126 nucleons have been found to be special, or magic, by having an exceptionally high natural abundance or being lighter than a gross trend would suggest¹⁶². The magic nucleon numbers were explained by the nuclear shell model^{163,164} as being numbers where nucleon shells were completely filled. The nuclei with such numbers of nucleons are referred to as (semi-)magic (a magic number of one type of nucleons) or doubly-magic (magic numbers of both nucleon types, such as ${}^4_2\text{He}$, ${}^{48}_{20}\text{Ca}$, or ${}^{208}_{82}\text{Pb}$, which is the heaviest known doubly-magic nucleus and at the same time the heaviest stable nuclide). Early calculations taking shell structure into account and extrapolating to the next shell closures led to the prediction that these would occur at $Z = 114$ and at $N = 184$ (refs¹⁶⁵⁻¹⁶⁷). The nuclei around this doubly-magic system were predicted to have lifetimes ranging from minutes to millions of years, and this had led to the notion of an ‘island of stability’^{165,168}. This island was far removed from the heaviest known nuclei, which by then were the members of the actinide series, that is with Z not higher than 103. This sparked searches for superheavy elements in nature¹⁶⁹⁻¹⁷¹ and in the laboratory. Although the latter has led to the synthesis of all elements up to element ${}_{118}\text{Og}$ (ref.⁵) the searches in nature occasionally led to discovery claims, but none was ever confirmed.

In the late 1990s, realistic models of nuclear structure were applied to superheavy nuclei and the very concept of the island was revised¹⁰. In particular, the pattern of nucleonic shells is expected to undergo significant changes due to the large Coulomb interaction¹⁷² and the large level density of single-nucleon states^{173,174}. Whereas the current nuclear models predict the shell-stabilized region of superheavy nuclei in the region of neutron numbers $N \approx 172 - 184$ and proton numbers $Z \approx 112 - 126$, the predicted lifetimes are significantly shorter than originally anticipated. The current situation is summarized in the figure. Common to virtually all modern predictions is the fact that stable or quasi-stable superheavy nuclei are not expected to exist. That is, whereas the influence of shell structure on lifetimes is well documented in the currently known heaviest nuclei, the island appears to be associated with ‘longevity’ rather than true ‘stability’. The determination of the island’s location and extension in the chart of nuclides remains a key question in nuclear physics.

The figure shows an excerpt of the chart of nuclei with the region of the heaviest elements (the inset plots some data from refs. ^{158,160,175,176}). The colored boxes denote experimentally known nuclei and their decay modes (green: spontaneous fission (SF); yellow: α -decay; brown: β^+ -decay/electron capture (EC); blue: β^- -decay). The gray band indicates the expected β -stability line¹⁵. Predicted spherical shell closures at $N = 172, 184$ and at $Z = 114, 120$ are marked in red. The inset shows experimental α -decay half-lives; they increase as a function of N thus supporting the concept of an island of enhanced stability. The pattern is similar for odd- Z elements and for the SF half-lives¹⁵⁸. The nuclei with the longest lifetimes obtained in several representative models (m1 (Ref.¹⁷⁷), m2 (Ref.¹⁷⁸), m3 (Ref.¹⁷⁹), m4 (Ref.¹⁸⁰), m5 (Ref.¹⁸¹), and m6 (Ref.¹⁸²)) are shown. There is no theoretical consensus about the location of the most stable nucleus, its half-life, and its dominant decay mode. The known heavy isotope ${}^{284}\text{Cn}$ that decays by SF is highlighted and may serve as an experimental benchmark; the calculated half-life of this isotope in the 6 selected models is also displayed, rounded to one order of magnitude. The predictions are scattered over 11 orders of magnitude. Other SF properties differ as well among different theoretical frameworks, as indicated by the predicted fission yields of ${}^{294}\text{Og}$ shown in Fig. 1.

Figure 1. The nuclear landscape. **(a)** The quantified chart of nuclides obtained in the Bayesian model averaging calculations using 11 nuclear global mass models, experimental separation energies, and information about nuclear existence⁴⁸. For every nucleus with $Z, N \geq 8$ and $Z \leq 119$, is shown the probability (marked by the color legend) that the nucleus is bound with respect to proton and neutron emission. The regions where nuclei have been experimentally observed and their separation energies have been measured are indicated by the yellow and red line, respectively. The valley of β -stability is marked by a dashed line. The question marks indicate the territory of superheavy nuclides at the limit of nuclear mass and charge, whose borders are unknown. **(b)** Expected exotic forms of nucleonic matter in superheavy nuclei. Solid black line shows the potential energy curve of the $^{466}_{156}$ nucleus obtained in relativistic *density functional theory* (DFT) calculations⁵⁰ versus the nuclear quadrupole deformation β_2 . Open red circles indicate selected points on this curve for which neutron density distribution cross sections are shown in the plane perpendicular (A,B) or along (C,E,F) the symmetry axis of the nucleus. The density profiles reflect their relative sizes with respect of the spherical shape in the minimum D at $\beta_2 = 0$. **(c)** Charge distribution of fission fragments from SF of ^{294}Og predicted in the hybrid model⁵⁴ (blue bands), Brownian shape-motion approach (dashed line)⁵⁵, and scission-point model (dash-dotted line)⁵⁶. Panel a is reproduced with permission from ref.⁴⁸, APS; panel b is reproduced with permission from ref.⁵⁰, APS; panel c is reproduced with permission from ref.⁵⁴, APS.

Figure 2. The main contribution to the ground state configurations of the superheavy elements by means of the DCBQ-CI2 method⁹³. The blue areas indicate occupied orbitals and the yellow ones represent partially filled orbitals. The placement of atoms within the periodic table of elements (PTE) is emphasized by the use of bold borders. The red boxes show the six elements, which cannot be placed unambiguously in the PTE⁹³.

Figure 3. An attempt to place the superheavy elements into the periodic table of elements (PTE) according to Fig. 2. Highlighted in red is element $Z = 145$, which is placed twice. The placement of elements $Z = 121, 122, 123, 124$ and 168 cannot be established unambiguously. In the 8th period, a few entries are doubly occupied, while others remain vacant. This highlights the breakdown of periodicity in the PTE for the heavier elements. The 15th entry in the f -block represents the first slot of the d -block which is left vacant to indicate the place of the f -block inserts.

Glossary terms

Nucleon drip line. The boundary beyond which atomic nuclei are unbound with respect to the emission of a nucleon.

Electron correlation. The difference between an atomic or molecular property evaluated with a single determinant at the Hartree-Fock level and the same evaluated using either a sum of determinants or many-body perturbation theory.

Rapid neutron-capture process (r-process). A network of nuclear reactions taking place in high-neutron density environments that is responsible for the creation of approximately half of the atomic nuclei heavier than iron.

Heavy ion-induced fusion reaction. Nuclear reaction induced by ions with $Z > 2$, with superheavy elements typically being produced in cold fusion reactions based on targets near ^{208}Pb that form a compound nucleus that is excited typically in the range of $E_{\text{CN}} < 20$ MeV and favourable for the production of the elements with $Z < 113$, or in hot fusion reactions using ^{48}Ca beams leading to more highly excited (hotter) compound nuclei which evaporate more neutrons to de-excite, favourable for the production of the elements with $Z > 112$.

Recoil separator. Electromagnetic separator isolating single superheavy nuclei from the intense primary ion beam and from unwanted byproducts of the nuclear formation reaction to allow their detailed study.

Multinucleon transfer reaction. Nuclear reaction in which beam nuclei and target nuclei exchange nucleons (in contrast to fusion reactions where they fuse, forming a compound nucleus comprising all nucleons of beam nucleus and target nucleus).

Radioactive-ion beam. Ion beam consisting of radioactive ions.

Bayesian machine learning. Machine learning methods based on/applying Bayesian statistics.

Coulomb frustration. The competition between the short-range attractive nuclear interaction and the long-range Coulomb repulsion leading to exotic topologies of nucleonic densities.

Relativistic effects. The difference between solutions of the nonrelativistic Schrödinger and the relativistic Dirac equation for a specific property.

Quantum electrodynamics. Relativistic quantum field theory of the interactions of charged particles with the quantized electromagnetic field (i.e., photons) containing in particular contributions of particle vacuum fluctuations (virtual electron-positron pairs) leading to the vacuum polarization and the electromagnetic field fluctuations leading to the self-energy.

Negative energy continuum. The continuum spectrum with energy below $-mc^2$ that are a solution to the Dirac equation.

Atom-at-a-time chemistry. Chemistry studies in which only single atoms of the element of interest are present due to small production rates and short half-lives.

Density functional theory. Density functional theory is an alternative to wave-function based methods using approximate functionals of the one-particle densities and currents.

Website summary: Advances in superheavy element studies providing insight into the nuclear and atomic structure and the chemical behavior of these exotic short-lived systems will help push to the limit of the periodic table of elements and revise the concept of the island of stability.