

Article

New Developments in the Production and Research of Actinide Elements

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Abstract: This article briefly reviews topics related to actinide research discussed at the virtual workshop *Atomic Structure of Actinides & Related Topics* organized by the University of Mainz, the Helmholtz Institute Mainz, and the GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany, and held on the 26–28 May 2021. It includes references to recent theoretical and experimental work on atomic structure and related topics, such as element production, access to nuclear properties, trace analysis, and medical applications.

Keywords: atomic structure theory; optical spectroscopy; actinides and transactinides; atomic and nuclear properties; thorium-229; trace analysis



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1. Introduction

The actinides refer to the series of elements at the bottom of the periodic table, which includes elements from actinium (Ac, $Z = 89$) to lawrencium (Lr, $Z = 103$). Apart from the current controversy about their positioning in the periodic table and about the composition of “Group 3” [1,2], the research on actinides is experiencing an upsurge with the development of sensitive and sophisticated spectroscopic techniques and regularly attracts attention in the scientific media. It is a multidisciplinary research area par excellence, encompassing experimental and theoretical atomic and nuclear physics, nuclear chemistry, and quantum chemistry, with links to astrophysics, nuclear medicine, nuclear forensics, trace analysis, and so on. A virtual workshop entitled *Atomic Structure of Actinides & Related Topics* was held on the 26–28 May 2021 and brought together many experts in the abovementioned areas to present and discuss their recent findings and outlooks. To commemorate this successful event, we have included the logo of the workshop in this article; see Figure 1. This current issue of the MDPI journal *Atoms* includes some of these scientific contributions for the same reason and in the hope of serving as a useful resource for future studies of the atomic structure of the heaviest elements. In this article we provide a brief overview of the topics presented at the workshop, focusing on aspects of atomic structure, without claiming completeness. For the various areas covered here, other review articles are available, to which we refer in the appropriate places.



Figure 1. Logo of the virtual workshop on the *Atomic Structure of Actinides & Related Topics*, 26–28 May 2021.

2. Element Production

The lack of availability and the radioactive nature of most actinide elements render their experimental investigation challenging. The lighter actinides up to uranium (U, $Z = 92$) are still found in the earth's crust today because they are either primordial, i.e., they were produced before the formation of the solar system and are not yet fully decayed due to their long half-lives, as is the case for ^{232}Th ($T_{1/2} = 1.405 \times 10^{10}$ a), and ^{238}U ($T_{1/2} = 4.468 \times 10^9$ a), or because they are continuously produced from the decay of the primordial ones [3]. The heavier actinides up to fermium can be produced in high flux reactors by breeding lighter actinide elements in successive chains of neutron capture reactions with subsequent β^- decays. Due to competing fission processes and the still-limited neutron flux, the quantities produced in such nuclear reactors become smaller and smaller with increasing atomic number. Although milligram quantities can still be obtained for californium (Cf, $Z = 98$), only microgram or even nanogram amounts are available (at most) for einsteinium (Es, $Z = 99$) and fermium (Fm, $Z = 100$), respectively [4,5]. The breeding chain ends as soon as the short-lived fission isotopes of the latter element are reached.

To produce elements beyond fermium, nuclear reaction mechanisms such as fusion evaporation reactions induced by accelerated heavy ion beams are exploited instead [6,7]. Often projectile currents in the range of one particle μA from large accelerator facilities are used in conjunction with thin-target production techniques. In this way, the heaviest actinides from mendelevium to lawrencium can be produced at rates of a few atoms per second at most, whereas heavier elements up to oganesson can be produced at much lower rates.

With multi-nucleon-transfer reactions at energies close or slightly above the Coulomb barrier, one may target the production of more exotic and rather neutron-rich isotopes of the heavy elements [8] at existing facilities such as the KISS facility at RIKEN, Japan [9], and the IGISOL facility at JYFL in Jyväskylä, Finland [10], or at upcoming ones such as the $N = 126$ -factory at ANL in Argonne, USA [11], and the NEXT project at the AGOR cyclotron facility in Groningen, Netherlands [7].

Far away from earth-based accelerators, different scenarios are conceivable as to how these elements could have been or are being produced in nature. Currently, neutron star mergers are considered to be the most promising candidates for the so-called rapid neutron capture process [3] for the production of such elements due to the enormous neutron fluxes involved. Detection would therefore require multimessenger astronomy and a detailed spectral analysis of the light coming from these distant sources. However, in order to interpret these processes in terms of element abundances and production yields through comparison with the latest astrophysical models, a profound knowledge of the spectral light emissions of the elements and thus their atomic structure is essential [12–15].

3. Atomic Structure Modeling

In the second half of the last century, many experimentalists and theorists devoted increasing attention to the study of the atomic structure of the actinides, which until then had been a poorly explored area. It has been recognized that relativistic effects play an important role in the theoretical description of these atomic species [16]. Therefore,

relativistic modeling of such atoms is indispensable today. Nevertheless, their accurate description remains a challenge, especially because they exhibit an open $5f$ shell, which provides space for a large number of valence electrons and thus access to numerous low-lying electron configurations. A prominent example of an actinide with a complex atomic structure is protactinium (Pa, $Z = 91$), for which a detailed analysis of its atomic level density already indicates quantum chaos behavior, as reported in [17,18]. Therefore, constraints on modeling these atomic systems are often direct implications of the greatly increased configuration space [19–22].

Meanwhile, various numerical approaches, such as multi-configuration Hartree–Dirac Fock (MCDHF), Fock-space coupled-cluster (FSCC), and configuration-interaction (CI), are constantly being developed to provide reliable predictions for ever more complex systems [22–24]. Only recently has progress been made in the *ab initio* framework, such that the atomic properties and spectra of actinium (Ac, $Z = 89$) [25], fermium (Fm, $Z = 100$) [26], mendelevium (Md, $Z = 101$) [27], lawrencium (Lr, $Z = 103$) [28–30], rutherfordium (Rf, $Z = 104$) [31,32], and dubnium (Db, $Z = 105$) [33] can be calculated with a relatively high degree of reliability.

These calculations guide and complement experimental studies of the atomic structure of the heavy actinides, as was accomplished several years ago for fermium [34,35] and nobelium (No, $Z = 102$) [36–41], and also predict the behavior of known superheavy elements that remain long out of the reach of experimental investigation, as is the case with oganesson (Og, $Z = 118$) [42–44]. In the recent years they have helped in pinning down the first ionization potential of the heaviest actinides that were measured utilizing surface ionization techniques [45,46] and in unveiling extraordinary possibilities for the realization of optical clocks based on highly charged actinide ions [47,48].

4. Experiments Targeting the Atomic Structure

Across the actinides, a wealth of information on their atomic structures can be obtained from databases such as NIST, albeit mainly for the lighter representatives of this series [49,50]. As the atomic number increases, the scarcity of information becomes apparent, not least because experimental investigations become increasingly complex and often tedious due to decreasing sample quantities. The methods used are often adapted to the element production process and optimized for studies on that specific element, and would usually need to be further developed to achieve higher sensitivity to cover other exotic or heavier radionuclides. We limit ourselves here to a few examples; for a detailed overview of some of these developments, the reader is referred to [51] and the references therein.

One example of contemporary experimental investigations of the heaviest actinides is the RADRIS technique. It is based on laser resonance ionization in a gas cell and was developed for atomic level searches on the element nobelium. The technique enabled the experimental observation and characterization of an optical transition of this element for the first time [41], which subsequently paved the way to extract the first ionization potential with high precision [52]. The RADRIS technique is constantly being optimized, with the focus now on spectroscopy of the next heavy element, lawrencium—the last member of the actinide series [53].

Another example is the novel method of laser resonance chromatography. Researchers have recently been proposed to extend laser spectroscopy to lawrencium and the refractory metals of the superheavy elements, enabling both atomic-level searches and subsequent high-resolution spectroscopy [54,55]. The method exploits optical pumping to change the transport properties of an ion as it drifts in helium gas, allowing optical resonances to be identified based on drift times alone, without using resonance ionization or fluorescence detection. A corresponding experimental setup is currently under construction [56]. Closely related to this, and due to the fact that transport properties are sensitive to electronic configurations, efforts are currently being devoted to making ion mobility spectrometry one of the useful tools for isobaric purification in actinide research [57].

5. Nuclear Properties

Another pertinent topic in this regard is the fact that optical spectroscopy can bridge atomic and nuclear physics. Because the interaction of the central nucleus with the atomic shell leads to subtle but measurable differences in the atomic line spectrum, hyperfine spectroscopy can be used to study nuclear properties such as nuclear electric and magnetic moments and changes in the mean square charge radii [58,59]. The question of which atomic transitions are most suitable for hyperfine spectroscopy depends essentially on the requirement that the sensitivity of the experimental method should not be greatly deteriorated and on the precision with which one can determine the nuclear parameters. Again, calculations based on atomic theory are often required, since some nuclear properties cannot be measured absolutely and—due to the scarcity of information—comparisons with isotopes already studied via other methods are only possible in particular cases.

Such studies are of paramount importance in the field of heavy elements. The fact that the actinides span from the closed spherical neutron shell with neutron number $N = 126$ to the deformed shell closure at $N = 152$ makes them an ideal experimental field for the study of shell effects and nuclear deformation, which are essential for understanding nuclear stability in the presence of the extreme repulsive Coulomb forces between the numerous protons in the nucleus.

Spectroscopically, interest in the study of actinides has increased in recent years due to the increasing sensitivity and selectivity of the applied techniques. Long-lived plutonium (Pu, $Z = 94$) isotopes were studied via laser spectroscopy in an atomic beam through resonance ionization spectroscopy and in a fast ion beam through collinear fluorescence techniques with high spectral resolution [60], and also for production in a buffer gas cell [61]. Actinium (Ac, $Z = 89$) was investigated via in-gas-jet, in-gas-cell, and hot cavity laser spectroscopy [62–65], whereas the latter technique could only recently be applied to californium (Cf, $Z = 98$) [66] and einsteinium (Es, $Z = 99$) isotopes [67].

For the study of even heavier elements, one may emphasize two examples. The RADRIS technique enabled experimental studies of $^{252-254}\text{No}$ -isotopes, which, using atomic theory, allowed, for instance, the extraction of the charge radii changes [68]. In addition to level searches on lawrencium, RADRIS experiments are continuing, targeting heavy actinides to reach more exotic radionuclides with half-lives ranging from below one second up to more than one hour [53], given that the atomic transitions are known.

The other example is the in-gas-jet laser spectroscopy. This technique exploits laser resonance ionization in a low-temperature and low-density environment of a collimated gas jet emerging from a buffer gas stopping cell to enable high spectral resolution and maintain a high sensitivity at the same time [65,69–71]. Setups using this technique are planned for the S^3 separator at GANIL, Caen [72–74]; the MARA separator at JYFL, Jyväskylä [75]; and at the SHIP separator at GSI, Darmstadt [76,77]. Since the extraction of sample radionuclides occurs relatively quickly, one may gain access to short-lived nuclear isomers such as the K -isomer of ^{254}No . With the development of the in-gas-jet technique, the required laser systems are being further developed [78–80] to accommodate the needs of high-resolution spectroscopy.

Other research in relation to heavy elements includes the study of higher-order deformations of nuclei [81] and studies of radioactive molecules [82], which are motivated by the search for new particle physics beyond the standard model.

For more details on laser spectroscopy, we refer the reader to recent reviews [58,59] in a broader context and to [51] in the context of actinide research.

6. The Thorium-229 Nuclear Isomer

Another topic of particular interest for fundamental research is the low-lying nuclear isomer of thorium-229, the existence of which was demonstrated experimentally a few years ago [83]. The isomeric state is only 8.28 eV [84,85] above the ground state of the nucleus and thus within the VUV range of optical probing. Due to this singular property, several applications regarding a nuclear clock based on optical transitions of this isomer

have been proposed [86], although several open questions concerning this isotope are still being investigated [87,88].

In many experiments the isomer is produced in the α -decay branch of ^{233}U . A promising new approach is currently being pursued to prepare the isomer online at increased efficiency via the β^- decay of ^{229}Ac [89]. In all experiments, however, the most important and urgent goal today is to determine the excitation energy with an accuracy that should allow much more precise spectroscopic studies in the future [90]. Since the energy of the nuclear isomer is in the range of atomic binding energies, internal conversion can dominate radiative decay, especially in a neutral atomic state [91,92]. This, along with the need for laser cooling in future applications, brings 2+ and 3+ cations of this isotope more into focus in this research field [93,94].

7. Trace Analysis and Medical Applications

As radionuclides of trans-uranium elements are artificially produced in nuclear reactors, their elemental and isotopic abundance in test samples depends on the fuel material and the neutron flux initially used. Therefore, quantitative analysis of these telltale isotopic abundances can be exploited to identify radioactive material and to determine the history of the sample for nuclear forensics, safeguards, and proliferation [95,96]. Some trace analysis techniques rely on laser resonance ionization or light absorption and therefore also require a thorough knowledge of atomic structure. For resonance ionization spectroscopy (RIS) of actinides, the development of efficient excitation and ionization schemes is ongoing [97–100], and recent reports attest to advances in the use of spatially resolved secondary neutral mass spectrometry combined with RIS on sample atoms [96,101,102].

Another technical application for actinide research arises from developments in targeted alpha-therapy, in which the locally released energy in alpha decay has been used for the treatment of cancer cells. For this medical application, alpha emitters such as ^{225}Ac are being studied in detail, which in turn requires an understanding of atomic properties, production and purification, and the underlying chemical behavior for selective binding to cancer cells [103,104].

8. Summary

The actinides in general, and the heaviest of them in particular, are very limited in availability. Because of this limitation and the lack of applications for most of them, even their basic physical properties have been scarcely studied. The increasing interest in the above-mentioned research aspects related to the atomic structure of actinides and the availability of increasingly sensitive spectroscopy techniques has recently pushed actinide research forward. Experimental and theoretical groups are jointly working on gradually filling in gaps in our knowledge regarding the atomic and nuclear properties of these fascinating elements at the bottom of the periodic table, so more exciting results can be expected in the near future.

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