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Stabilizing nuclear shell effect in the heaviest elements may weaken with distance from stability line

Protons, Neutrons, Atoms, Elements... The Basic

Matter around us consists of roughly 90 elements, which are contained in the Periodic Table of the elements. Every atom consists of a small atomic nucleus, which comprises almost the total mass of the atom, and an extended electron shell. The nucleus itself is built up from (positively charged) protons and (neutral) neutrons, whereas the shell consists of (negatively charged) electrons.

To form a neutral atom, the number of positrons and electrons has to be identical (in case of different numbers, we speak of ions, which are then charged, instead of atoms). The chemical properties of an atom are determined by the number of electrons in its shell, and hence the number of protons in its nucleus, which is its "atomic number" (Z), according to which elements are ordered in the periodic table. Atoms can bind together, forming, e.g., molecules, or crystals.

Atomic nuclei with the same number of protons, i.e., from the same element, are called isotopes of this element.

The heavier an atom gets, the more protons are contained in its nucleus. With increasing proton number, the repulsive force of these protons will eventually lead to immediate disintegration of the nucleus. A topic of intense research concerns the question of the heaviest possible element. To date, all elements up to element 112 as well as elements 114 and 116 are officially recognized as discovered, and there are reports about the observation also of element 113, 115, 117, and 118 published. It is currently not clear, which element is the heaviest one that can exist.

Nuclear shell structure - The "Island of Stability" of Superheavy Elements

While the elements beyond uranium typically become more short-lived with increasing atomic number, decade-old predictions based on the nuclear shell model suggest this trend to be broken and even reversed, once a next "magic number" is approached. Such magic numbers originate from proton- and neutron shells that are completely filled at these numbers. Filled (or: closed) shells render the corresponding nuclei to be more stable that non-magic ones, and exhibit a spherical shape (while other nuclei are often deformed). The heaviest confirmed magic proton number for spherical nuclei is 82, corresponding to the element lead (Pb). The heaviest confirmed magic neutron number for spherical nuclei is 126. The nucleus ^{208}Pb, having 82 protons and 126 neutrons, is thus "doubly magic" and strongly stabilized by the nuclear shell effects.

Traditionally, the next spherical magic number has been 114, but different theoretical models differ with some favoring 114, while other prefer 120 or even 126. There is more consensus on the next spherical neutron number, which is expected to be 184. At and around these next shell closures, much more stable nuclei are expected to occur compared to those presently known (which are all still at least 7 neutrons away from N=184). The shell

effects most directly affect the tendency to fission spontaneously into lighter elements and delay this process by huge factors, probably millions of millions of time. This lead to the term of the "Island of Stability of Superheavy Elements" and has triggered many searches for such superheavy nuclei in nature. However, the superheavy nuclei known today decay mostly by emission of an alpha-particle: obviously this decay mode is less delayed by the shell-effects than that of spontaneous fission. Whether the most long-lived superheavy nuclei decay by spontaneous fission or by alpha decay, and what the longest half-lives will be, is not yet firmly known. Indications for the existence of the "Island of Stability" come from the observation that different isotopes of, e.g., copernicium or flerovium become more long-lived, the more neutrons their nuclei contain. The isotope Cn-285, for example, has a half-life of about 30 seconds, while the isotope Cn-277, having 8 fewer neutrons, decays with a half-life of about 0.5 micro seconds (60 million times faster!).

Unfortunately, the direct tracing of the strength of the nuclear shell effects in elements around Z=114, or even at Z=120 or higher, is not currently possible due to the very small rates at which, e.g., Z=114 atoms can be produced and detected of just a few atoms per day. Therefore, theoretical calculations are used to provide our knowledge on the strength of these effects. These models can be tested in more accessible regions of the chart of nuclei. For example, available data on the evolution of this shell closure up to Z=91 indicate this to remain strong at least up to that proton number, i.e., the nucleus ^{217}Pa. By studying the nuclei ^{221}U (which has N=129 and was discovered in our experiment) and ^{222}U(with N=130, and for which no detailed data on its nuclear decay properties were known before our experiment), we were able to study the region around N=126 in the Z=92 (i.e., uranium) isotopic chain and to extract indications that hint at a weakening of this shell closure in the region that far away from stable territory around ^{208}Pb. Theoretical models can thus be tested against our new data, which is expected to help refining them to be able to yield more reliable data also in the region of the island of stability.

To detect the decays of the very short-lived ^{221}U (half-life: 660 ns) and ^{222}U (half-life: 4.7 ms), we used the Combined ANalog and DIgital (CANDI) data acquisition system, see below *The new CANDI data acquisition system*.

Experiment Facts and Figures

In the experiment, an intense beam of ^{50}Ti, which is readily available at GSI in high intensity over extended periods was used to irradiate targets of ^{176}Yb (which is a stable isotope of ytterbium) produced at the GSI target laboratory. The target wheel system is described in the article "High intensity target wheel at TASCA: target wheel control system and target monitoring" by Egon Jäger et al. Over the course of the experiments, the UNILAC accelerator at GSI delivered $1.6 \cdot 10^{17}$ beam particles of titanium-50.

The analysis of the data revealed twenty-six decays of the new isotope ^{221}U and eighty-one decays of ^{222}U , which for the first time allowed measuring the α-decay energy of this latter isotope, of which only three decays were observed in an experiment performed in 1983, but did not have a fast data acquisition system available and was hence unable to separate the decay signal of ^{222}U from that of its fast-following daughter ^{218}Th.

How TASCA Works

In collisions of $1.6 \cdot 10^{17}$ projectiles of the titanium-50 beam with the $1.5 \cdot 10^{18}$ ytterbium-176 atoms per cm^2 contained in the targets, many different nuclei are produced from different types of nuclear reactions. Many - in fact most - projectiles even leave the target unchanged. The observation of the about 100 atoms of interest thus requires a very good separation from all other, unwanted nuclei.
After the target, all products enter a region of 0.8 mbar helium and collide with the helium atoms. In the collisions, electrons are transferred to the reaction products. All products of a certain kind, e.g., all element 117 ions (yes, ions, they are charged!) soon end up in the same charge state, regardless of the charge state at which they left the berkelium-249 target. The ions then enter the region of the magnetic field of the TASCA dipole magnet, which bends their trajectory due to the Lorentz force. As they all have the same mass, roughly the same velocity, and the same charge state, their trajectories are identical and they are guided into the direction of the focal plane of the separator. Species other than element 117 take different turns and will miss the focal plane.
After the dipole magnet, two quadrupole magnets act like a lens and focus the beam of element 117 into an area in the focal plane that is small enough that a sensitive detector can be placed there.

In the TASCA focal plane, the ions penetrate a gas-ionization chamber, where they leave some energy, which is measured, and which indicates that an ion passed through. They then implant into a silicon detector, which is 144 x 48 mm^2 large, where they are stopped. This implantation is registered and triggers the clock for the measurement of the lifetime of the implanted ion. As the detector consists of about 7000 individual pixels of 1 x 1 mm^2 area, the position at which the ions implant is determined with a precision of 1 mm. If an implantation event indeed is from a radioactive isotope (like the uranium ones), the subsequent decay, e.g., by emission of an alpha particle, is also registered in the silicon detector. Decay events can easily be distinguished from implantation events due to the absence or presence of a signal from the gas-ionization chamber. The silicon detector allows measuring the time differences between different events, and hence the lifetimes of the observed nuclei, and the energies released in the decay. Our system was improved to be sensitive also for very short-lived nuclei, where events occur with only a very small time difference, see below *The new CANDI data acquisition system*.

The new CANDI Data Acquisition System

For the detection of charged particles like α particles or fission fragments, semiconductor Si detectors are used. The energy brought into the detectors is converted into charge, which is collected, amplified, and shaped into an (analog) pulse signal. This is then digitized, in which form it is suitable for further processing. Classical data acquisition systems use peak sensitive analog-to-digital converters.
Such electronics have been successfully operated for decades and for many experimental purposes. There are some drawbacks connected to such systems, though: one is that the shaping of the signal, which needs some time. In case a next even occurs during this time, the energy from this event might add to the energy of the event being processed, which leads to a summed signal which looks like it originates from a higher-energy event than was actually occurring. The exact energy will depend on when exactly the second event occurred relative to the start of the shaping process; as this cannot be determined, no information on

the energies of the two summed events can be obtained - in fact it will not even be possible to determine that the resulting event is the (partial) sum of two lower-energy signals. Another issue is the time that the system needs to digitize the signal and store it; this can be as long as several microseconds, or even more. During this time, the system is busy and unable to accept a next event to be processed: it is temporarily "dead" (leading to the term "deadtime" for this time).

As TASCA was used also for the search and study of superheavy nuclei, which potentially have half-lives shorter than few microseconds, the detection and data acquisition system was upgraded in 2011. While one branch of the system operates as outlined above, a second, digital branch of electronics modules was developed at the GSI Experiment Electronics department. This branch takes snapshots of the signal shape in an early phase of the singal processing chain, i.e., traces the processes happening in the Si-detector. By setting the length of the so recorded traces to a long enough time before and after the occurrence of an event, this digital branch is able to fully cover the deadtime of the analog branch, i.e., while this is busy. The trace length in our experiment was selected to be 50 µs. Thus, CANDI overcomes both above mentioned problems of more standard electronics. In case that two events occur within such a short time that they sum up in the analog branch, the trace in the digital branch will show this. Also, if an event occurs during the deadtime of the analog branch, this will also be shown in the traces from the digital branch.

The lower limit for the half-life of isotopes that can be studied at TASCA is now no longer limited by the data acquisition system, but is given by the flight-time of the reaction products from the target, through the separator, into the detector, which is about 0.7 microseconds. CANDI is faster than this, and the minimum time difference of events happening in the silicon detector that can still be resolved, e.g., decays of daughters of implanted nuclei, is about 100 ns.

Related Articles:

I. Rusanov et al.
FPGA Hit Finder and Energy Filter for the FEBEX Pipelining ADC
GSI Scientific Report 2013

J.Hoffmann et al.
New TASCA Data Acquisition Hardware Development for the Search of Element 119 and 120
GSI Scientific Report 2011

Our Experiment - What's New?

The main aspects of our experiment are the following:

The new isotope ^{221}U

The existence of the uranium isotopes with mass numbers 216 to 219 (neutron numbers 124 to 127) and from 222 (neutron number 131) onward has been established before our studies. Our observation of ^{221}U is thus contributing to closing the gap, leaving only ^{222}U unknown in this region. Based on the observation of 26 decays, we established that ^{221}U

decays by emission of α particles with an energy of 9.71±0.05 MeV, with a half-life of 660±140 ns.

New and improved data on ^{222}U

The existence of ^{222}U was known before our work. However, the only experiment observing this isotope was performed in 1983, using instrumentation that was state of the art back then, and reported the observation of the decay of three atoms. Only lifetimes could be measured, but not the energies of the emitted α particles. Based on three measured lifetimes, the half-life was estimated to be around 1 ms. However, also the information on the α-particle energy is needed to get access to information on the strength of the N=126 shell closure in uranium. In our experiment, 81 decays of ^{222}U were observed. The energy of the emitted α particles was measured to be 9.31±0.05 MeV and the half-life was 4.7±0.7 μs.

Evolution of the N=126 spherical shell closure

From the available data on uranium isotopes around N=126, including our new data, we could calculate for different isotopes the probability for the pre-formation of an α particle inside a nucleus that is subsequently emitted in an α decay. It is known that magic nuclei, which are more tightly bound than non-magic ones, have a reduced probability for this pre-formation of an α particle, compared to nuclei without magic numbers. Combining this data for the uranium sequence, especially a comparison of the isotopes with N=126 and those with N=130, with the corresponding data of isotopes with the same neutron number for elements with smaller Z, we find that the influence of the magic N=126 neutron number is smaller in uranium compared to its influence in lighter elements with even Z between 82 and 90. This may hint at a weakening of the stabilizing effect of the N=126 shell closure, hence at its washing-out in uranium.

That the strength of the N=126 shell closure is weakening can also be deduced from the energy gap between the last neutron filling up the N=126 shell and the first neutron beyond this shell. Different families of theoretical models predict how the strength of this shell effect should develop when moving away from the magic Z=82 (i.e., doubly-magic ^{208}Pb). A comparison of the experimental data with a model representative of a whole family of models, in which the atomic nucleus is described like a macroscopic charged nuclear liquid droplet, whose properties are slightly modified by shell effects, reveals this model to perform well at and near Z=82, it fails to describe the decreasing trend observed experimentally when going towards uranium. On the other hand a model representative for a family, in which the properties of a nucleus are computed by considering the interactions between its individual nucleons, does a better job describing the evolution of the N=126 shell strength, but deviates significantly in the region of ^{208}Pb. Looping back our findings into refining theoretical models will thus help to improve these, thus having direct impact towards more reliable predictions, e.g., on the evolution also of the next shell closures in the region of the island of stability of superheavy elements.

What about the still missing ^{220}U?

The 220U with N=128 is still missing after our experiments. Its a decay populates the N=126 isotope ^{216}Th. Due to the magic neutron number, this is significantly more stable. The ^{220}U therefore decays at an accelerated rate, meaning that it will be the most short-lived isotope

of the sequence. In fact, its lifetime is expected to be significantly shorter than the flight-time through TASCA (see below *How TASCA works*), meaning that its direct production and identification is not feasible. Therefore, an indirect production pathway appears more promising. This involves production of the (unknown) ^{224}Pu isotope with N=128, which would likely undergo α decay to ^{220}U. As this decay does not feed a magic isotope, it will not be accelerated, and the expected lifetime of 224Pu is long enough for it to survive the flight through TASCA into the focal plane detector, where its decay could be observed thanks to CANDI. However, as ^{224}Pu has two extra protons compared to uranium, its production rate will be small, so an experiment designed to identify ^{224}Pu will need significant amounts of accelerator beamtime.

Practical Implications?

NO !

Given the production and/or observation rate of just a few atoms per hour, and their fleeting lifetimes of the order of microseconds, no practical implications are imminent.

The Scientific Article

The New Short-Lived Isotope ^{221}U and the Mass-Surface Near N = 126 was submitted on July 14, 2015 and published in **Physical Review Letters** on 10 December 2015. (Phys. Rev. Lett. 115, 242502 (2015))

The first author is Dr. Jadambaa Khuyagbaatar from the Helmholtz Institute Mainz, a joint daughter of the GSI Helmholtzzentrum in Darmstadt and the University Mainz. He is a core person of the local Darmstadt/Mainz superheavy element group and was responsible for tuning the nuclear reaction, for a part of the TASCA separator and detection system, the data acquisition, analysis, and interpretation, and wrote the paper (naturally with input from the co-authors).

The Collaboration

Our collaboration comprises 39 scientists and engineers from 8 institutions in 6 countries:

- Helmholtz Institute Mainz (HIM), Mainz, Germany
- GSI Helmholzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany
- Johannes Gutenberg University Mainz, Mainz, Germany
- University of Liverpool, Liverpool, United Kingdom
- Lund University, Lund, Sweden
- Saha Institute of Nuclear Physics, Kolkata, India
- University of Jyväskylä, Jyväskylä, Finland
- Institute for Electron Technology, Warsaw, Poland

The local superheavy element group from Darmstadt/Mainz working on this experiment comprises:

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- Michael Block (GSI Darmstadt)
- Holger Brand (GSI Darmstadt)
- Christoph Düllmann (University of Mainz, GSI Darmstadt, and Helmholtz Institute Mainz)
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- Fritz Peter Heßberger (GSI Darmstadt and Helmholtz Institute Mainz)
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