May 01, 2014

Superheavy Element 117 Confirmed - On the Way to the "Island of Stability"

The Scientific Article

Study of the ^{48}Ca + ^{249}Bk Fusion Reaction Leading to Element Z=117: Long-Lived α-Decaying ^{270}Db and Discovery of ^{266}Lr
was submitted on February 22, 2014 and published in Physical Review Letters on May 01, 2014
(Phys. Rev. Lett. 112, 172501 (2014))
Synopsis: Element Z=117 Confirmed

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 of the element 117 collaboration, was responsible for tuning the nuclear reaction, for a part of the TASCA separator and detection system, the data acquisition and analysis, and wrote the paper (naturally with input from the co-authors).

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.

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).

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!).

Searches and studies for ever more stable superheavies keep going on.

Experiment Facts and Figures

Berkelium-249 decays with a half-life of 330 days, predominantly by beta-minus decay to californium-249. Due to this relatively short half-life, experiments with berkelium-249 require excellent coordination between Oak Ridge National Laboratory, which is the only center worldwide, which can produce this isotope, and the experimental facilities where it is used in scientific experiments, such as the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany.

The berkelium-249 target material produced at Oak Ridge National Laboratory

The start of the experiment dates back to March 2010, when the production of irradiation targets at the Oak Ridge National Laboratory's High Flux Isotope Reactor (HFIR) began. These targets, containing the transuranium element curium (atomic number 96), were irradiated in the HFIR's intense neutron field for about 16 months. Atomic nuclei of curium capture neutron, becoming beta-minus unstable and decaying into berkelium (atomic number 97), producing the exotic radioisotope berkelium-249 (Bk-249). The irradiation ended in August 2011. After a cooling period, the target processing to extract the produced Bk-249 was carried out at ORNL's Radiochemical Engineering Development Center (REDC) and about 25 mg of Bk-249 were obtained, representing the total world production. Half of the material was delivered to the Flerov Laboratory for Nuclear Research, a research center in Dubna, Russia. The other half was shipped to the Institute for Nuclear Chemistry at the Johannes Gutenberg University Mainz, Germany on February 27, 2012.

Target production at the Institute of Nuclear Chemistry, University of Mainz

On March 06, 2012, the Bk-249 arrived at the Institute for Nuclear Chemistry at the Johannes Gutenberg University Mainz, where the facilities and the expertise to convert the material into a target suitable to withstand the intense heavy-ion beam from the GSI accelerator facility are available. To prevent the material to overheat under the beam power, it is deposited on a wheel-like structure, which rotates through the beam, thus ensuring dissipation of the heat over maximum possible area. To this end, four banana-shaped segments, each covering 90° of the wheel, were produced. The material was electrochemically deposited on 2-micrometer thick titanium foils in the form of Bk-2O3. The production process was highly efficient with a chemical yield of >95% of the material received from ORNL. The individual segments were delivered to the GSI Darmstadt on March 23, 2012. Production of the targets is described in the article " Preparation of actinide targets for the synthesis of the heaviest elements" by Jörg Runke et al.

The 2012 berkelium-target campaign at the TASCA separator at GSI Darmstadt

Upon arrival, final preparations were performed and the segments were mounted onto the target wheel. 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.
Initially, the target was irradiated with a titanium-50 beam, which leads to the new element with atomic number 119 upon fusion with the berkelium-249. The element 119 campaign, starting on April 14, 2012, lasted for four and a half months. The analysis of the data from the element 119 search experiment is still ongoing.

On September 26, 2012, irradiation of the berkelium-249 target material was resumed, but the beam was changed from titanium-50 to calcium-48. The fusion of calcium-48 (element 20) with berkelium-249 (element 97) produces atomic nuclei of element 117, as was previously reported from experiments conducted at the research center in Dubna, Russia. The irradiation lasted for about one month, ending on October 29, 2012.
The UNILAC accelerator at GSI delivered 10^{19} beam particles of the stable but rare isotope calcium-48.

The analysis of the data revealed four correlated alpha decay chains of two different types. The present paper focuses on two of these four, which are of very similar nature and originate from the same isotope, most likely 117-294, i.e., the nucleus with 117 protons and 177 neutrons. The two chains are mostly in agreement with decay chains previously published from the Flerov Laboratory for Nuclear Research in Dubna, Russia. A new finding, though, concerns the late members of the decay chain. The seventh member of the decay chain, dubnium-270, was previously reported to decay exclusively by spontaneous fission, i.e., to split into two lighter nuclei. Instead, we have observed dubnium-270 to undergo alpha-decay in both cases, thereby populating the new isotope lawrencium-266, which subsequently decayed by spontaneous fission with a half-life of the order of 11 hours, see

also *What's New*?.
The "hay stack" of non-reacting primary calcium particles and vast number of unwanted nuclear reaction byproducts was efficiently separated from the element 117 "needles" by means of the gas-filled recoil-separator TASCA. The possibility to detect the alpha-decay of dubnium-270 is owed to the high sensitivity of the TASCA separator and detection system, see also *How TASCA works*.

The isotope 117-294 has a half-life of about 50 milliseconds - or between 35 and 92 milliseconds accounting for statistical uncertainties.

How TASCA Works

In collisions of projectiles of the calcium-48 beam with berkelium-249 target atoms, many different nuclei are produced. Many - in fact most - projectiles even leave the target unchanged. The observation of two atoms in one month requires a very good separation of all unwanted nuclei and selective detection of those of interest: those from element 117. 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 a signal, indicating 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 element 117 ones), the subsequent decay, e.g., by emission of an alpha particle, or a spontaneous fission, 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.

Classical detection systems operating with so-called "analog" electronics need about 30 microseconds to process a measured event. During this time, they cannot register additional events, they are 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 much shorter than 30 microseconds, the detection and data acquisition system was recently upgraded and a second, digital branch of electronics modules was developed at the GSI Experiment Electronics department. This branch takes snapshots of everything that happens in a pixel, while the other, analog branch is dead. Hence, events happening during the deadtime are registered, which enables experiments also with extremely short-lived nuclei. The lower limit for the half-life of isotopes that can be studied is now given by the flight time of the reaction products from the target, through the separator, into the detector, which is about 0.7 microseconds. The system is also ideally suited to register unwanted reaction products that decay very quickly, with lower energies than superheavy nuclei. In purely analog systems, these energies sum up to an artificial total energy, which is unrelated to the decay of any single nucleus, but may end up in the energy region where decays of superheavy nuclei occur. Hence, TASCA allows to very selectively discriminate between unwanted nuclear reaction products (the few that are not deflected fully in the magnetic separator and still end up in the focal plane detector) and true high-energy alpha-decays (typically above 10 megaelectronvolts, MeV) from superheavy nuclei.

Thanks to this advanced system, which suppresses background from unwanted products, TASCA is suitable to measure also comparatively long-lived isotopes. Only this allowed the safe identification of the new alpha-branch in dubnium-270, which went unregistered in previous studies.

Our Experiment - What's New?

The main aspects of our experiment are the following:

Independent observation of element 117

True, there are earlier reports on the observation of element 117. In 2010 and in 2012, collaborations working at the Flerov Laboaratory for Nuclear Reactions in Dubna reported data using the same nuclear reaction, claiming the same isotopes as reported in our paper. In both experiments, they used the same device. So while this may make our result sound boring at first, an independent verification of an important discovery announcement is of paramount importance in science. It is also an explicit requirement by IUPAC to officially accept a new element as "discovered" (see also *What is the Procedure to Name a New Element?*). There are many cases in the history of science where a discovery was claimed, but follow-up studies, often more sensitive ones, were unable to confirm the original finding (see also *Does Element 117 Occur Naturally?*). This may be due to experimental flaws, due to malfunctioning of the experimental equipment, due to misinterpretation of the obtained data, to name but a few possibilities. Our reports therefore strengthen the case for element 117 significantly: the team was largely different, as was the used setup.

Observation of new new decay mode in dubnium-270 and discovery of the new isotope lawrencium-266

All four decay chains assigned to 117-294 reported from the Flerov Laboratory for Nuclear Research in Dubna comprised six a particles and were then terminated by a spontaneous fission after 33, 38, 24, and 1.2 h. The a particles were assigned to the decays of 117-294, 115-290, 113-286, Rg-282, Mt-278, and Bh-274. The spontaneous fission was thus ascribed to Db-270. In the three first decay chains, some a particles were observed between the Bh-274 a decay and the spontaneous fission, but they could not be assigned to a specific isotope, because the rate of such events in the detector was too large to exclude them to be of random origin from byproducts of the nuclear reaction, unrelated to the element 117 decay chains. Only in the last chain, no such particles were observed. In the preparations for our experiment, we upgraded the detection and data acquisition system to distribute the rate of events reaching the focal plane of TASCA over almost 7000 pixels, which leads to a very small rate of events in one individual pixel. Additionally, thanks to the digital data acquisition branch (see *How TASCA works*), we can clearly distinguish between true a particles and the summing of two low-energy a particles (originating from lighter elements than comprised in the element 117 decay chain) occurring within a very short time. This allowed the safe identification of a further a particle in each of the two chains, originating from the previously unobserved a decay of Db-270. The spontaneous fission terminating the chain thus originates from Lr-266 in our chains, and most likely also in the three decay chains from Dubna, where the spontaneous fission occurred about one day after the last a decay. As such, the finding of a new decay mode in Db-270 may again sound to be of minor importance. The impact for superheavy element research is due to the fact that with a setup like the upgraded TASCA separator, the safe measurement of such long-lived nuclei decaying by a decay is now possible at all! The closer the center of the "Island of Stability" is approached, the longer-lived the nuclei become. Finding such isotopes is one of the main driving forces behind superheavy element research. The caveat is that - as long as single nuclei can only be registered when they decay - it becomes harder and harder to measure the decay of nuclei that are ever less prone to decay in the first place. The demonstration that nuclei with half-lives up to about one day can now safely be measured, regardless of whether they decay by a decay or spontaneous fission (the two main decay modes registered so far for such heavy nuclei, see *The "island of stability" of superheavy elements*) is a big step forward for the exploration of the heaviest elements.

Alternative approaches, like storing of single ions of superheavy elements in ion traps hold the promise to allow the identification of even more long-lived isotopes. However, the sensitivity of such setups needs to be boosted to reach the sensitivity to safely measure ions produced at a rate of single events per week or even month. At GSI, the SHIPTRAP setup is ideally suited for such experiments, see M. Block et al. in Nature 463, 785 (2010) and E. Minaya Ramirez et al. in Science 337, 1207 (2012).

Is Element 117 a Halogen or a Metal?

Naturally, element 117 is placed in group 17 of the Periodic Table of the elements (often also called the 7th main group), which contains the halogens. It finds its place just below astatine with its 85 protons. When going down along the members of group 17, the elements are getting more and more metallic, with astatine being a semi-metal and no longer a typical halogen like the lighter members of that group. If this trend were followed, element 117 would likely be a rather volatile metal. Fully relativistic calculations agree with this expectation, however, they are in need of experimental confirmation. In fact nuclear chemists have developed methods to determine basic chemical properties (e.g. gas or metal?) based on the measurements of single atoms - and the TASCA separator at GSI was built for that purpose.

Most recently, a study on the chemical properties of flerovium (element 114) found this element to be a volatile metal, see the publication "Superheavy element flerovium (element 114) is a volatile metal" by Alexander Yakushev (who is a key person also for the current element 117 experiment) and co-authors.
Such single-atom chemical studies require the element of interest to live sufficiently long,

with the current limit being on the order of about 1 second. The two known isotopes of element 117 are both too short-lived. However, in the decay chains reported in the current publication, all lighter members from elements 115, 113, … down to lawrencium (element 103) are more long-lived and may be chemically studied in future experiments

To produce longer-lived isotopes of the superheavy elements, isotopes containing more neutrons need to be synthesized. These are then located closer to the next "magic" neutron number 184. In the future, there might be means to produce such more neutron-rich isotopes.

Does 117 Occur Naturally?

In past decades, the finding of very long-lived superheavy elements in nature has been claimed repeatedly. However, none of these searches were verified in independent experiments to date. In contrast, recent "verification attempts" were often significantly more sensitive than the experiments leading to the original claims, but were negative. Recent examples can be viewed at

- Physical Review C 85, 024315 (2012)
- Physical Review C 83, 015801 (2011)

On the other hand, there are long-standing theoretical predictions that superheavy elements might be a product of stellar supernova explosions, there synthesized by means of the rapid neutron-capture process. See, for example, Nature 231, 103 - 106 (1971)

The question concerning the endpoint of the rapid neutron-capture process is still open, so the question cannot currently be answered with certainty. However, even if produced, most likely even the anticipated most long-lived nuclei near the "Island of Stability" are too short-lived to be observed in the solar system, namely on Earth.

Practical Implications?

NO !

Given the production and/or observation rate (two atoms per month), any practical implications are seemingly far-fetched.

Where is "The End" - How Many Superheavies Can We Create and Observe?

First of all: Yes! There are more "new elements" in reach.

Evidence for elements up to 118 has been reported by Yu.Ts. Oganessian and his collaborations from experiments conducted at Dubna. Calcium-48 (20 protons, 28 neutrons) was and is being used as beam, impinging on target foils comprising various actinide elements ranging from uranium (proton number 92) up to californium (proton number 98). 20+98=118.

Elements beyond californium cannot not be produced in amounts large enough to produce a target suitable for an accelerator experiment. Therefore, the production of elements beyond 118 requires abandoning the doubly-magic calcium-48 as a beam and use, instead, a beam of a heavier element. One option could be titanium-50, as it was used, e.g., in the search experiment on element 119 (see Experiment Facts and Figures). Other options that were already used in attempts to produce element 120 involve beams of chromium (element 24), iron (element 26), or nickel (element 28).

There are two major problems though:

> *First of all,* the production probability of the fused systems (e.g., with Z=120) decreases rapidly. So instead of observing up to one or two atoms of these superheavy elements per day, as was the case in the element 115 experiment, or two per month observed for element 117, this rate will decrease to a handful per year with current setups. While such experiments are in principle feasible, as evidenced by the TASCA element 119 search, such very long around-the-clock experiments are very demanding on both material and personnel!
> To increase the production rate for currently known elements, which will allow their detailed study, and to get access to new elements, work towards the construction of a new accelerator at GSI Darmstadt has started. Led by the ACcelerator design and Integrated Detectors (ACID) research section of the Helmholtz Institute Mainz, a first part of the new accelerator is currently under construction.
>
> *Secondly,* the lifetimes of the superheavy nuclei which can be produced with a given selection of stable ion beams (calcium, titanium, chromium, iron etc.) and actinide target materials become shorter and shorter. Eventually, they may not survive the flight path through the separators, i.e. never make it to the detection set-up! Maybe their decay daughters can be observed, but that poses severe constraints on the discovery. While TASCA for example can register nuclei if they live longer than 0.7 microseconds only, the development of yet faster systems will become more and more important. Alternatively, in the future, more neutron-rich, hence radioactive ion beams may allow the synthesis of more neutron-rich isotopes than are accessible with stable beams. These may then be long-lived enough for observation in separators as are already available.

Beyond nuclei residing on the island of stability, even heavier nuclear systems with exotic properties are predicted to exist. These include hyperheavy nuclei with hollow nuclei, forming "bubbles", or of toroidal shape!

Ultimately, there are the neutron stars - ultradense extended nuclear matter.

The TASCA Element 117 Collaboration

A part of the TASCA collaboration presents data on element 117 at GSI Darmstadt
Photo: G. Otto / GSI

Our collaboration comprises 72 scientists and engineers from 17 institutions in 10 countries:

- Helmholtz Institute Mainz (HIM), Mainz, Germany
- GSI Helmholzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany
- Johannes Gutenberg University Mainz, Mainz, Germany
- Advanced Science Research Center, Tokai, Ibaraki, Japan
- Oak Ridge National Laboratory (ORNL), Oak Ridge, TN, USA
- University of Liverpool, Liverpool, United Kingdom
- Australian National University, Canberra, Australia
- Lund University, Lund, Sweden
- Lawrence Berkeley National Laboratory (LBNL), Berkeley, CA, USA
- Lawrence Livermore National Laboratory (LLNL), Livermore, CA, USA
- Vanderbilt University, Nashville, TN, USA
- Saha Institute of Nuclear Physics, Kolkata, India
- University of Oslo, Oslo, Norway
- University of Jyväskylä, Jyväskylä, Finland
- Paul Scherrer Institute, Villigen, Switzerland
- University of Bern, Bern, Switzerland
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