

EVIDENCE FOR THE POSSIBLE EXISTENCE OF A LONG-LIVED SUPERHEAVY NUCLEUS WITH ATOMIC MASS NUMBER $A = 292$ AND ATOMIC NUMBER $Z \cong 122$ IN NATURAL Th

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Evidence for the possible existence of a superheavy nucleus with atomic mass number $A = 292$ and abundance of about 1×10^{-12} relative to ^{232}Th has been found in a study of natural Th using inductively coupled plasma-sector field mass spectrometry. The measured mass is different from any species with molecular mass of 292, but it matches the predictions for the mass of an isotope with atomic number $Z = 122$ or a nearby element. Its deduced half-life of $t_{1/2} \geq 10^8$ y suggests that a long-lived isomeric state exists in this isotope. The possibility that it might belong to a new class of long-lived high spin super- and hyperdeformed isomeric states is discussed.

Keywords: Superheavy elements; mass measurements; natural Th; $A = 292$; $Z \cong 122$; isomeric states; superdeformation; hyperdeformation.

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1. Introduction

The question “how heavy can a nucleus be” is a fundamental problem in nuclear physics.¹ Experimentally, elements up to $Z = 118$ have been produced synthetically by heavy-ion reactions^{2,3} with the half-lives of the $Z = 106$ –118 nuclei ranging from a few minutes to about a ms. However, in a recent study of natural Th substances,⁴ long-lived isomeric states have been observed in the neutron-deficient $^{211,213,217,218}\text{Th}$ nuclei. Since they were found in natural Th the deduced lower limit of their half-lives are $t_{1/2} \geq 10^8$ y, or otherwise they would have decayed away. This half-life is 16–22 orders of magnitude longer than their corresponding g.s. (A

long-lived isomeric state with a half-life of $>1.2 \times 10^{15}$ y is known in ^{180}Ta .⁵) It has been hypothesized⁴ that they might belong to the recently discovered⁶⁻⁹ class of long-lived high spin isomeric states in the second minimum (superdeformed (SD) minimum)^{10,11} and/or in the third minimum (hyperdeformed (HD) minimum)¹²⁻¹⁴ of the nuclear potential energy when plotted as a function of deformation. This result motivated us to search for long-lived superactinide elements in natural materials. According to the extended periodic table,¹⁵ the superactinides are classified as homologues of the actinides, with elements 122 and 124 placed as eka-Th and eka-U, respectively. As a working hypothesis, it is reasonable to assume that if any of the superactinide elements exists in nature they might be carried along with Th and/or U substances. Here we report on evidence for the possible existence of a nucleus with atomic mass number 292 that was found in natural Th with an estimated half-life of $t_{1/2} \geq 10^8$ y and an abundance of about 1×10^{-12} relative to ^{232}Th . Its measured mass is different from the mass of any potential species with molecular mass of 292, but it matches the predictions^{16,17} for the mass of an isotope with atomic number $Z = 122$ and also some isobars of neighboring elements. Based on theoretical chemical predictions¹⁸ it is argued that it is probably element 122. Preliminary results of this work have been presented previously.¹⁹

The mass M_A of an atom is equal to:

$$M_A = ZM_H + NM_n - BE, \quad (1)$$

where M_H and M_n are the masses of the hydrogen atom and the neutron, respectively, and BE is the binding energy of the nucleus. The binding energy per nucleon (BE/u) of stable nuclei has a broad maximum around $A \approx 60$, with a value of 8.7 MeV/u, which falls monotonically to about 7.6 MeV/u at Th and U.²⁰ The predicted BE/u values for the superactinide nuclei are around 6.9 MeV/u.^{16,17,21} Therefore, masses of superactinide isotopes are higher than, and resolvable from, the masses of all molecules with the same mass number, except for multihydrogen-containing molecules. This is seen in Fig. 1, where the systematic behavior of the

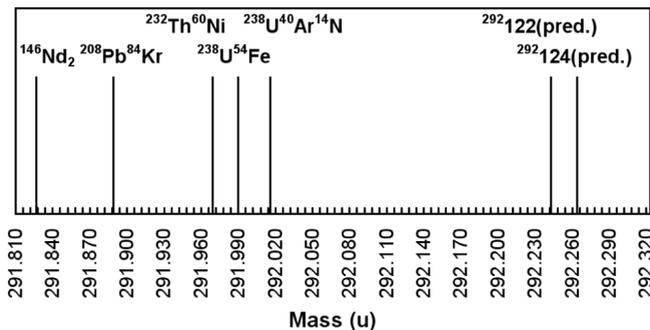


Fig. 1. Representation of the systematic behavior of the masses of various $M = 292$ species, ranging from the symmetric combination $^{146}\text{Nd}_2$ to Pb-, Th- and U-based molecules,²² to the predicted^{16,17} masses of $^{292}122$ and $^{292}124$ nuclei.

masses²² of various $M = 292$ species, from the symmetric combination $^{146}\text{Nd}_2$ to Pb-, Th- and U-based molecules to the predicted^{16,17} masses of the $^{292}122$ and $^{292}124$ nuclei are displayed. The masses of the very neutron-rich ^{292}Th and ^{292}U nuclei are predicted to be 292.399 and 292.362 u,²¹ values well above the expected masses of the $^{292}122$ and $^{292}124$ nuclei, that are 292.243 and 292.264 u, respectively.^{16,17} (The calculated masses of these two nuclei in Refs. 16, 17 agree to within 0.001 u.) Thus, as was demonstrated before,²³ accurate mass measurements are an effective tool in searching for naturally occurring superheavy elements.

In principle, natural minerals like monazite, which is the common source of Th, are expected to be the most promising materials to study. However, spectral background was the main obstacle when looking for isotopes with relative abundance of $(1-10) \times 10^{-11}$ in natural materials.⁴ Therefore, purified natural Th was used in our measurements. Another advantage of using purified Th is that the chemical properties of Th are known.

In the present work, we performed accurate mass measurements for masses 287–294 in Th solutions. Evidence was obtained for the possible existence of an isotope with a mass that matches the predictions for atomic mass number 292 and Z around 122. Here we describe this observation.

2. Experimental Procedure

The experimental procedure was similar to that described earlier.⁴ Inductively coupled plasma-sector field mass spectrometry (ICP-SFMS) was used for the experiments. The ICP-SFMS is an Element2 (Finnigan, Thermo-Electron, Bremen, Germany). In this instrument, a solution of the material to be studied is introduced into a high temperature (6000–8000 K) plasma source. At these temperatures, predominantly atomic species are present. Molecular ions are produced after the source, mainly by interaction with oxygen and hydrogen ions from the solution. The predefined medium resolution mode, $m/\Delta m = 4000$ (10% valley definition), was used throughout to separate atomic ions from molecules with the same mass number. The sensitivity-enhanced setup of the instrument was similar to that described in Ref. 24. It provided sensitivity for ^{232}Th in this resolution mode of up to 2×10^8 counts $\text{s}^{-1} \text{mg}^{-1} \text{l}^{-1}$. The sample uptake rate was 60–80 $\mu\text{l min}^{-1}$. Methane gas was added to the plasma to decrease the formation of molecular ions.²⁵ Oxide and hydride formation (monitored as UO^+/U^+ and UH^+/U^+ intensity ratios) were approximately 0.04 and 1×10^{-5} , respectively. In its standard mode of operation, the measured masses in the ICP-SFMS are limited to $M \leq 270$. In order to measure higher masses, the accelerating voltage was reduced from 8 to 7 kV. The machine was calibrated using the ^{230}Th , ^{235}U , ^{238}U , $^{238}\text{U}^{16}\text{O}$ ($M = 254$), and $^{238}\text{U}^{40}\text{Ar}^{16}\text{O}$ ($M = 294$) molecules. Then a new calibration was performed using the $^{232}\text{Th}^{40}\text{Ar}^{16}\text{O}$ ($M = 288$) and $^{238}\text{U}^{40}\text{Ar}^{16}\text{O}$ molecules. This calibration was checked regularly every half to one hour of the experiments and the data were corrected accordingly. During the measurements, the magnetic field was kept constant and the scanning was done by changing the accelerating voltage.

1000 mg l⁻¹ purified Th solutions were purchased from two companies. The solutions were analyzed in three sessions: 25 February (run I), 25 May (run II), and 5 July (run III), 2007. A range of about 0.42 u, divided into 60 channels, was scanned in each measured spectrum in the first two runs, and a range of about 0.2 u, divided into 30 channels, was scanned in the third run. During the first session, masses from 287 to 294 were measured using the Th solutions with an integration time of 1 s channel⁻¹, and these measurements were made three to five times each. In one spectrum, two events were observed with a solution produced by Customer Grade (from LGC Promochem AB, Borås, Sweden) at a mass that fits the predictions for atomic mass number 292 and a *Z* value around 122. This mass region was again studied in runs II and III with the same solution.

3. Results

Complete elemental screening was performed on the Th solution to assess the impurity concentration levels. The concentrations of certain trace elements that could potentially give rise to spectrally interfering molecular species, expressed as ppm of the Th concentration, are as follows:

U 80, Bi < 0.1, Pb 0.2, Hg < 0.1, Au < 0.1, Os < 0.1, Hf < 0.1, Dy 0.7, Nd 3, Ce 0.7, La 0.4, Ba 0.2, Cs 0.2, I 1.0, Te < 0.1, Fe 7, Ni 3, Ca 50, K 1000, B 3, Be < 0.1.

Instrumental sensitivity could vary between runs as a result of matrix effects caused by the introduction of highly concentrated solutions into the ICP source. In order to minimize this effect the inlet of the machine was cleaned before each run. It was shown before⁴ that a concentration of about 50 mg l⁻¹ of Th gave a correct peak position and good resolution. Therefore, our measurements were begun with this concentration. During the first run, the Customer Grade Th solution, diluted to 50 mg Th l⁻¹ of 0.7 M HNO₃, was studied. Figure 2 shows the results in the mass region 288 where the peak of the ²³²Th⁴⁰Ar¹⁶O molecule is observed. The

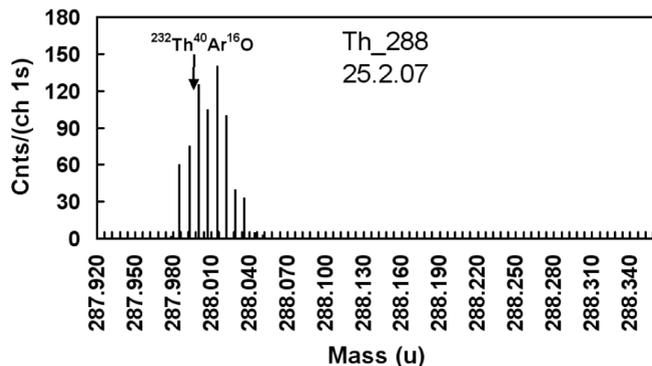


Fig. 2. Results of mass measurements for mass region 288. The c.m. of the observed peak is 288.009 u and the known mass²² of ²³²Th⁴⁰Ar¹⁶O is 287.995 u (position marked by arrow).

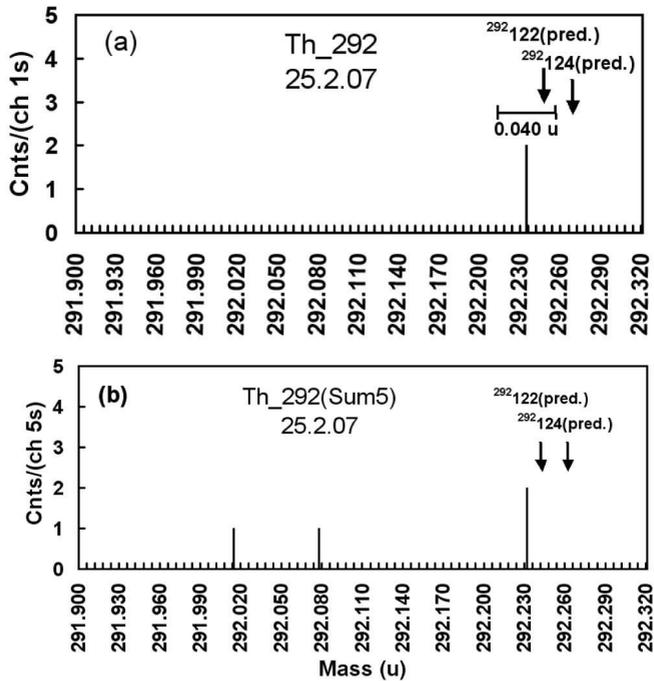


Fig. 3. Results of measurements for mass region 292 obtained in the first run. A single spectrum out of five measured is shown in (a). The sum of the five spectra is displayed in (b). The arrows indicate the positions of the predicted^{16,17} masses of the $^{292}_{122}$ [$^{292}_{122}\text{(pred.)}$] and $^{292}_{124}$ [$^{292}_{124}\text{(pred.)}$] isotopes.

center of mass (c.m.) of this peak is shifted from the known mass of this molecule²² by 0.014 u. The full width at half maximum of this peak is about 0.040 u. This resolution is somewhat lower than obtained before⁴ of 0.030 u, due to the decrease of the accelerating voltage to 7 kV. It is, however, adequate for our experiments since, as seen in Fig. 1, the mass difference between the heaviest possible molecule with a mass number of 292 and the predicted masses of the superactide atoms is about 0.220 u.

The results obtained in run I for the mass region of 292 are presented in Fig. 3. Figure 3(a) shows a spectrum where the integration time per channel was 1 s. The two events seen in this spectrum are at mass 292.232 u. (A correction of -0.014 u due to the calibration shift mentioned above was applied in deducing the mass.) The observed mass is close to the predicted values of the $^{292}_{122}$ and $^{292}_{124}$ nuclei as indicated by the two arrows in this figure.

Figure 3(b) shows the sum of five spectra, including the spectrum seen in Fig. 3(a). As can be seen, only two background events at lower mass were added in the additional four spectra.

In run II, mass 292 was scanned 60 times with the same 50 mg l⁻¹ Th solution as in run I. Figure 4(a) displays the sum of three spectra out of the 60 measured.

In addition to the $^{238}\text{U}^{40}\text{Ar}^{14}\text{N}$ peak, four events are seen in the vicinity of the predicted masses of the isotopes $^{292}122$ and $^{292}124$. Figure 4(b) presents the sum of the 60 spectra. Compared with Fig. 4(a), two events were added in the region from 292.130 to 292.320 u in the additional 57 spectra. This shows that the four events around mass 292.260 u are not background, since if they were background events one expects about 80 counts in this region in Fig. 4(b). (The reason that this group has been observed in only a few spectra out of the 60 measured is probably because of its low abundance. For a solution concentration of 50 mg l^{-1} , an abundance of 1×10^{-10} will result in 1 count s^{-1} on average. A lower abundance of about 5×10^{-12} will result in one event every 20 s, or about one measurement in 20.) Figure 4(c) shows the sum of the 60 spectra measured with a blank solution of 0.7 M HNO_3 . Only two events are seen in the whole region from 292.020 to 292.320 u. The c.m. of the high mass peak seen in Figs. 4(a) and (b) is 292.272 u, taking into account a correction of 0.013 u deduced from the measured shift of the $^{238}\text{U}^{40}\text{Ar}^{14}\text{N}$ peak seen in Fig. 4(a).

In run III, the concentration of the Th was increased to 80 mg l^{-1} . In this run, we focused on the region between 292.130 and 292.320 u (denoted by the small upward arrows on the X-scale in Fig. 4(b)), which was divided to 30 channels and was scanned 200 times. Each spectrum was measured with an integration time of 1 s channel^{-1} . The calibration was determined before and after every 100 spectra using the $^{232}\text{Th}^{40}\text{Ar}^{16}\text{O}$ ($M = 288$) and $^{238}\text{U}^{40}\text{Ar}^{16}\text{O}$ ($M = 294$) peaks. The sum spectrum of the 200 scans is seen in Fig. 5(a). An equivalent spectrum with a blank solution is seen in Fig. 5(b). An accumulation of 36 events is seen in Fig. 5(a) at a c.m. position of 292.263 u. The background in the same region in Fig. 5(b) is 7 counts. The background in an equivalent region from 292.154 to 292.226 u in Fig. 5(a) is 14 counts. (These counts could be due to the tail from the $^{238}\text{U}^{40}\text{Ar}^{14}\text{N}$ peak seen in Fig. 4.) Taking into account the average of these two backgrounds, the net number of counts is 25 ± 7 . The error was estimated according to the formula $\sigma = \sqrt{(N_t + N_b)}$, where N_t is the total number of counts and N_b is the number of background counts.²⁶

The weighted average of the mass of the newly observed peak seen in the three runs is $292.262 \pm 0.030 \text{ u}$. The intensity ratios of this peak among the three runs is 25:4:2. This is in accord, within statistical accuracy, with the expected ratio of 25:4.7:0.4. The latter values were calculated by normalization to the results of run III, account taken of the different times and solution concentrations used in the measurements. It is estimated that the concentration of the species responsible for this peak is about 1×10^{-12} of ^{232}Th , or about 1×10^{-16} of the solution.

Statistical analysis, done in the same way as described earlier,⁴ shows that the probability that the observed 292.262 u peak is due to an accidental concentration of events is about 3×10^{-5} , for each of the three measurements separately.

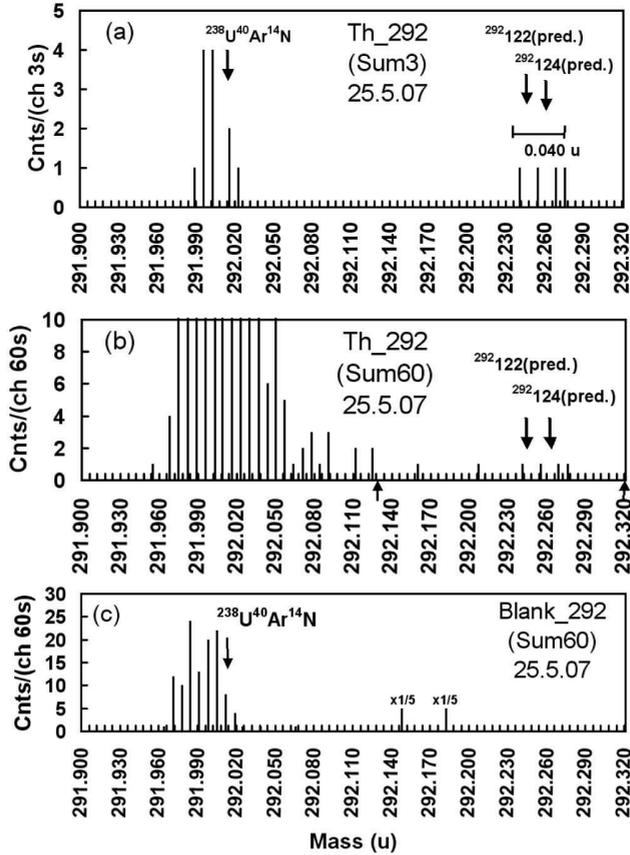


Fig. 4. Results of measurements for mass region 292 obtained in the second run. The sum of three spectra out of the 60 measured is shown in (a), and the sum of the 60 spectra is shown in (b). The two rightmost arrows indicate the positions of the predicted^{16,17} masses of the $^{292}122$ [$^{292}122(\text{pred.})$] and $^{292}124$ [$^{292}124(\text{pred.})$] isotopes. The left-hand arrows (in (a) and (c)) indicate the known position²² of $^{238}\text{U}^{40}\text{Ar}^{14}\text{N}$. The c.m. of this peak is 292.003 u and its known mass is 292.016 u. The small upward pointing arrows on the X-scale in (b) at masses 292.130 and 292.320 u indicate the limits chosen for the third run. The sum of 60 spectra of the blank solution is shown in (c). The events denoted by $\times 1/5$ have one count each.

4. Discussion

We were unable to match the signals of the suspected superactinide isotope with any molecular ion. As described in the introduction and displayed in Fig. 1, because of binding energy, masses of molecular aggregates are lower than the mass of the aggregates. As already mentioned, the mass of the very neutron-rich ^{292}Th nucleus is predicted to be 292.399 u,²¹ well above the measured peak.

Another possibility that needs to be considered is the potential presence of hydrocarbon-based molecular ions from pump oil. However, no contamination from pump oil was seen in the spectra of the blank.

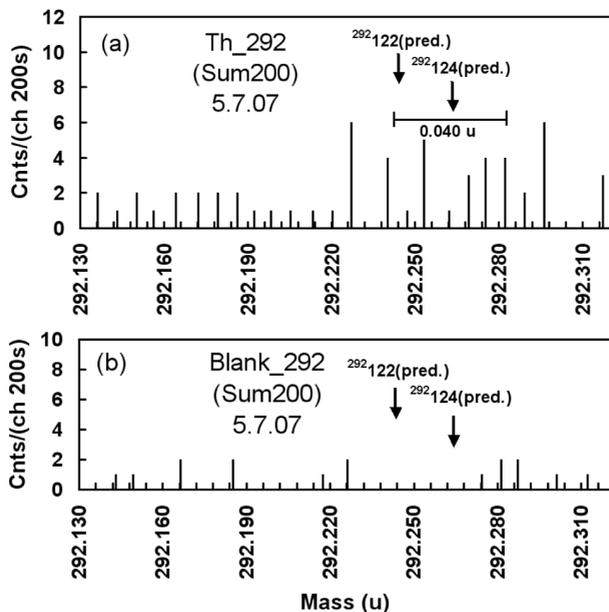


Fig. 5. Results of measurements for mass region 292 obtained in the third run. The sum of 200 spectra in the limited mass region of 292.130 to 292.320 u (corresponding to the upward arrows on the X-scale in Fig. 4(b)) is shown in (a). Figure 5(b) shows the sum of 200 blank solution spectra. The arrows indicate the positions of the predicted masses of the $^{292}_{122}$ [$^{292}_{122}(\text{pred.})$] and $^{292}_{124}$ [$^{292}_{124}(\text{pred.})$] isotopes.^{16,17}

At the same time, the measured mass fits the predictions for the atomic masses of superactinide isotopes with $A = 292$ and Z values from 121 to 126,^{16,17} which vary monotonically from 292.236 to 292.291 u, respectively. The prediction for isotope $A = 292$ of element 122 (eka-Th) is 292.243 u. The atomic configuration of Th is $6d_{3/2}^2 7s^2$ and its chemical separation is based on its stable 4^+ oxidation state. The predicted configuration of element 122 is $8s^2 7d_{3/2} 8p_{1/2}$.¹⁸ In spite of the different configuration, element 122 is also expected to form a stable 4^+ oxidation state. Element 121 has only three electrons outside the filled shells of element 118 (eka-Rn). It seems unlikely that it would form a stable 4^+ oxidation state. Elements above $Z = 122$ have more electrons outside the closed shells, but their configurations have not been rigorously calculated. One can assume that they might form a 4^+ state but also higher oxidation states. If element 122 exists in nature together with Th, it is reasonable to assume that it followed Th in the chemical separation and showed up in our measurements. However, the possibility that the observed $A = 292$ isotope belongs to an element of somewhat higher Z cannot be excluded.

The predicted half-lives of the normal g.s. of nuclei around $^{292}_{122}$ are of the order of 10^{-8} s.²⁷ This suggests that the observed events are due to a long-lived isomeric state in $^{292}_{122}$ or in a nearby isobar. If its initial terrestrial concentration was similar to that of ^{232}Th , then the lower limit on its half-life would be about

10^8 y, or otherwise it would have decayed away. The character of the observed isomer was not measured directly. It cannot be related to the actinide fission isomers, since their lifetimes are in the region of ns to ms. As mentioned in Ref. 4, it is probably not a normal high spin K-type isomer,²⁸ because the lifetimes of all known K-isomeric states in neutron-deficient nuclei with $Z \geq 84$ are not longer than several minutes. It could be a high spin isomer near doubly closed shells. In addition to the recurrent predictions that $Z = 114$ and/or 126 and $N = 184$ could be the next proton and neutron closed shells, respectively,¹ isotopes of $Z = 120$ and $N \cong 184$ and $N = 172$ have also been predicted recently to be doubly closed shell nuclei.²⁹⁻³² The Q_α value of $^{292}122$ is predicted to be about 14 MeV.^{16,17} It is estimated that in order to have a half-life of about 10^8 y for such high value of Q_α , $\Delta L_\alpha \approx 30\hbar$. Such a high spin difference is unlikely. One could hypothesize, as we did in the case of the Th isomers,⁴ that the proposed state is an aligned high spin SD or HD state,⁶⁻⁹ where the high spin, the barriers between the various minima of the potential-energy surfaces and the radioactive decay properties contribute to the long lifetime. It was also pointed out⁴ that for high spin states produced by alignment, the barrier against fission increases significantly³³ due to specialization energy.³⁴

High spin states in general, and in SD and HD minima in particular, are made preferentially by heavy-ion reactions.^{9,35} If the observed state in $A = 292$ turns out to be of the high spin SD or HD type, then heavy-ion reactions could be involved in its nucleosynthesis.

5. Summary

In summary, mass spectral evidence has been obtained for the possible existence of a long-lived superheavy isotope with an atomic mass number of 292 and $t_{1/2} \geq 10^8$ y. Based on predicted chemical properties of element 122, it is probable that the isotope is $^{292}122$, but a somewhat higher Z cannot absolutely be excluded. Because of its long lifetime, it is deduced that the observed isotope at $A = 292$ is in a long-lived isomeric state rather than in the g.s. The hypothesis that it is a high spin SD or HD isomeric state is discussed.

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