Verification of Filling-Up of 5f Electrons and Confirmation of the Actinide Series by Determination of the First Ionization Potentials of Fm, Md, No, and Lr

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The first ionization potential (IP_1) of an atom is one of the most fundamental chemical and physical quantities of every element, which gives direct information about the binding energy of an electron in the outermost electronic orbital of an atom. Accurate IP_1 values of heavy elements provide crucial tests for our understanding of their electronic structure affected by strong relativistic effects caused by their large electric charge of heavy nuclei. IP_1 values of heavy elements with atomic number $Z \geq 100$, however, could not be determined experimentally, because their production rates and half-lives drastically decrease as the atomic number increases. The study of these elements therefore requires new techniques on an atom-at-a-time scale.

Recently, the successful measurements of IP_1 of lawrencium (Lr) and nobelium (No) in an atom-at-a-time scale have been reported [1, 2]. Those were measured by using a method based on surface ionization coupled to mass separation and α-particle detection techniques, and a laser resonance ionization spectroscopy, respectively. However, a systematic IP_1 measurement from fermium (Fm) to Lr has not been performed.

In this work, we have applied the surface-ionization method [1] to determine the IP_1 values of Fm, mendelevium (Md), and No. In addition, IP_1 of Lr has been also measured to improve the accuracy of the previously reported IP_1. Surface ionization process takes place on a solid surface kept at a high temperature and can be described by the Saha−Langmuir equation. The ionization efficiency (I_{eff}) depends on the work function of the ionizing material, ϕ (eV), the temperature of the material surface, T (K), and IP_1 of the element. Short-lived isotopes ^{249}Fm ($T_{1/2}$ = 2.6 min), ^{251}Md ($T_{1/2}$ = 4.27 min), ^{257}No ($T_{1/2}$ = 24.5 s), and ^{256}Lr ($T_{1/2}$ = 27 s) were produced in nuclear fusion reactions of actinide targets with heavy ion beams. The produced atoms recoiling from the target were transported to a surface ion-source installed at the JAEA-ISOL (Isotope Separator On-Line) by the He/CdI_2 gas-jet transport system. Transported products were surface ionized on the surface of the Ta cavity kept at temperatures between 2550 and 3000 K in the ion source. Produced ions were extracted and mass separated in the ISOL. The number of collected ions after the mass separation was determined by α spectrometry to calculate I_{eff} values. Measured I_{eff} values for each isotope gave IP_1 values of the elements. The obtained IP_1 values of Fm, Md, No and Lr were determined to be 6.52 ± 0.13, 6.59 ± 0.13, $6.62^{+0.06}_{-0.07}$, and $4.96^{+0.05}_{-0.04}$ eV, respectively.

The variation of the IP_1 values of the heavy actinides with atomic number in comparison with those of the heavy lanthanides is shown in Figure 1. The measured IP_1 values agree well with those predicted by state-of-the-art relativistic calculations performed alongside the present measurements [3]. As expected from the prediction, the IP_1 values increase up to No via Fm and Md with atomic number in analogy to the heavy lanthanides. We take this as an indication that the 5f orbital is fully filled at No with the $[Rn]5f^{14}7s^2$ configuration. The lowest IP_1 value of Lr is confirmed; the ground-state electronic configuration of the Lr atom has closed $5f^{14}$ and $7s^2$ shells with an additional weakly bound electron in the valence orbital. This IP_1 structure is similar to that found in heavy lanthanides, confirming unambiguously that the actinide series end with Lr.

References

[1] T. K. Sato et al., Nature **520**, 209-211(2015).
[2] P. Chhetri et al., Phys. Rev. Lett. **120**, 263003 (2018).
[3] T. K. Sato et al., J. Am. Chem. Soc. **140**, 14609-14613(2018).
[4] A.Kramida et al., NIST Atomic Spectra Database, https://physics.nist.gov/asd

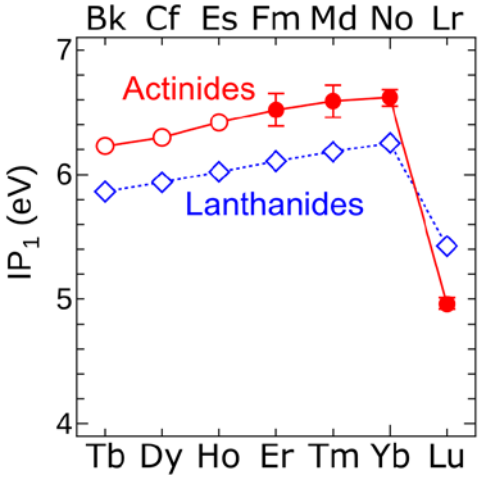

Fig.1 The first ionization potentials of heavy lanthanides (blue) and actinides (red) including the present results for Fm, Md, No and Lr. A closed symbol indicates the data obtained in this work. Open symbols are taken from literature values [4]