Measurement of the First Ionization Potential of Lawrencium (Lr, Z=103) by a Surface Ionization Method

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Relativistic effects influence the electronic structure of heavy elements. The ground-state electronic configuration of the heaviest actinide, lawrencium (Lr, Z = 103), is predicted to be [Rn]$5f^{14}7s^{2}7p_{1/2}$, which is different from that of the lanthanide homolog lutetium (Lu) [Xe]$4f^{14}6s^{2}5d$. The reason for this change is that the 7p orbital of Lr is stabilized below the 6d orbital by strong relativistic effects [1].

The first ionization potential (IP_1), one of the most fundamental physical and chemical properties of an element, 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. IP_1 values of heavy elements with $Z \geqq 100$, however, could not be determined experimentally, because production rates drastically decrease for these elements as their atomic number increases. The study of these elements therefore requires new techniques, on an atom-at-a-time scale.

In order to determine the IP_1 value of Lr experimentally, we employ the surface ionization process in which an atom is ionized via interaction with a solid surface at a high temperature. For this purpose, we have developed a surface ion-source installed to the JAEA-ISOL system and an efficient detection system for α-particle measurements [2]. In this work, we carried out a measurement of the IP_1 value of Lr based on the dependence of the ionization efficiency (I_{eff}) on IP_1 in the surface ionization process.

The isotope ^{256}Lr ($T_{1/2}$ = 27 s), produced in the reaction ^{249}Cf(^{11}B, 4n), was used for studying the Lr ionization. A ^{249}Cf target (thickness 260 μg cm^{-2}) was irradiated with a 67.9-MeV ^{11}B beam from the JAEA tandem accelerator. To obtain a relationship between I_{eff} and IP_1 in our present system, short-lived ^{142,143}Eu, ^{143}Sm, ^{148}Tb, ^{153,154}Ho, ^{157}Er, ^{162}Tm, ^{165}Yb, ^{168}Lu and ^{80}Rb were also produced by irradiating mixed targets with a ^{11}B beam. The reaction products recoiling from the targets were transported to the surface ion-source by a He/CdI_2 gas-jet transport system [3]. The products ionized on a tantalum surface of the ion-source cavity were extracted, accelerated with 30 kV, and mass-separated. The ionization experiments were performed at the ion-source temperatures of 2800 and 2700 K. The amount of ions collected at the end of the ISOL was determined with an α-particle detection system. The I_{eff} was evaluated by comparing detection rates of ionized ^{256}Lr to the rate at which ^{256}Lr transported directly from the target to the detection system were registered.

By using the present system, we successfully ionized and mass-separated ^{256}Lr with efficiencies of 36 ± 7% and 33 ± 4% at 2800 K and 2700 K, respectively. From these I_{eff} values of Lr, the Lr IP_1 value was determined to be $4.96^{+0.08}_{-0.07}$ eV using the relationship between I_{eff} and IP_1 obtained by the I_{eff} measurements of the short-lived isotopes.

A theoretical calculation of the IP_1 of Lr was also performed, using the relativistic coupled cluster approach with single, double, and perturbative triple excitations (DC CCSD(T)), and corrected for the Breit contribution and Lamb shift. The calculated IP_1 for Lr is 4.963(15) eV. Our experimental result is in excellent agreement with the theoretical value (Fig. 1) [4].

Thus, we have experimentally shown that the IP_1 of Lr is distinctly lower than that of Lu (5.425871(12) eV). Lr, the heaviest actinide element, has the lowest IP_1 value of all lanthanides and actinides; this quantitatively reflects and confirms the theoretically predicted situation of closed $5f^{14}$ and $7s^2$ shells with an additional weakly-bound electron in the valence orbital. We note that the surface ionization method, successfully applied here to determine the IP_1 of Lr, can provide experimental data that can benchmark quantum chemical calculations of the heaviest elements.

References

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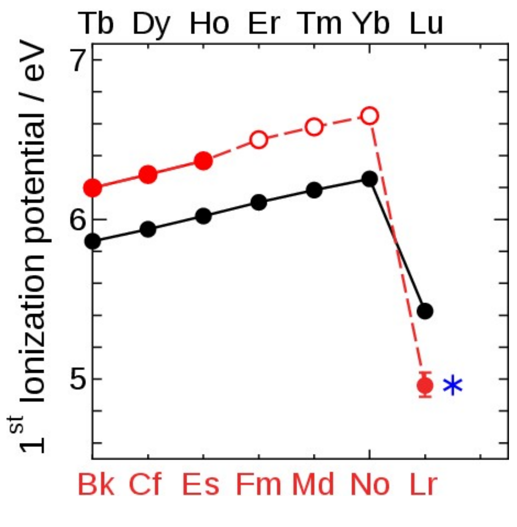

Fig.1 Ionization potential of heavy lanthanides (black symbol) and actinides (red symbol) including our present results for Lr. A closed and open symbol indicates an experimental and estimated value, respectively. The blue star * depicts the present theoretical value for Lr.