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Development of relativistic density functional theory with finite-light-speed correction toward electronic structures of super-heavy elements

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Chemical properties of trans-uranium and super-heavy elements, i.e., electronic properties in their isolated atomic form, are interesting objects of research as well as the nuclear properties. The ionization energy of lawrencium ($Z = 103$) was recently measured to be $4.96\,\mathrm{eV}$ at the tandem accelerator at the Japan Atomic Energy Agency (JAEA) [1]. This result and the ionization energy of lutetium, which is the elements above lawrencium in the periodic table, show a non-monotonic dependence among the other f-block elements. Moreover, the tendency of the ionization energy of lutetium and that of lawrencium are also different from each other. After that, the council of the International Union of Pure and Applied Chemistry (IUPAC) began to discuss whether lutetium and lawrencium belong to d-block elements or f-block elements[2]. Nevertheless, the origin of the different behavior of their ionization energies has not been understood.

To understand the origin of the tendency of such chemical properties, relativistic effects for the electronic structure are thought to be important. Pyykkö calculated the stable electronic configurations and proposed a periodic table of elements theoretically up to $Z = 172$ by using the Dirac Hartree-Fock equation with the Coulomb interaction [3]. However, the anomaly of the ionization energy of lawrencium mentioned above is not yielded according to his calculation. Several years later, Pershina calculated the ionization energy, electron affinity, and absorption enthalpy mainly by the coupled cluster single, double, and perturbative triple (CCSD(T)) method [4-7]. Although these quantities were accurately evaluated by theoretical calculations, the mechanism of the anomaly was not discussed.

To calculate the properties of the trans-uranium and super-heavy elements, we develop the density functional theory (DFT) with the relativistic effects. Since the numerical cost of DFT is much less than the wave function methods, such as the coupled-cluster method, DFT can be easily applied to molecules and solids as well. In our work [8], finite light-speed effect in $O\left(1/c^2\right)$, i.e., the Breit correction [9], is considered as well as effects incorporated by the use of the Dirac equation instead of the Schrödinger equation.

With the conventional non-relativistic calculation, the electronic configurations of the d-block elements are stabilized for both lutetium and lawrencium. In contrast, the present relativistic correction stabilizes the p-block configuration in lawrencium, while the d-block configuration is kept stable for lutetium. This difference of the electron configurations may cause the anomaly of the ionization energy of lawrencium. Since the numerical cost of our method is lower than the wave function methods, our method enables us to study chemical and physical properties of super-heavy elements systematically.

Our method is also promising for calculation of other fundamental physical properties such as the electric dipole moment of electrons.

In this talk, we briefly introduce the idea of our method, its results including the possible origin of the anomaly of the ionization energy of lawrencium, and future perspectives.

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