HIGH-ACCURACY CALCULATIONS FOR HEAVY AND SUPERHEAVY ELEMENTS

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The relativistic coupled-cluster method starts from the Dirac-Coulomb-Breit Hamiltonian,

$$H_{DCB} = \sum_i h_D(i) + \sum_{i<j} \left(\frac{1}{r_{ij}} + B_{ij} \right)$$

where h_D is the Dirac one-electron operator with a finite, uniformly-charged nucleus, and B_{ij} is the unretarded (or frequency-independent) Breit interaction. The self-consistent four-component Dirac-Fock-Breit orbitals are calculated, and correlation is included by the Fock-space coupled-cluster method. The method has been applied over the last five years to a large number of heavy elements, with the main objects of interest being transition energies (ionization potentials, excitation energies, electron affinities). Many transition energies are calculated for each atoms, with results usually within 0.1 eV of experiment. The success of RCC has made it possible to use the method as a predictive tool for the super-heavy elements (Z $>$ 100), where experimental data is scarce.

The talk will start with a brief review of the method; sample applications will then be presented. Comparison with experimental data will be shown for some heavy elements. A recent development is the hermitian coupled-cluster method, which yielded significantly improved values for group-13 elements: the np ionization potentials and ${}^2P_{3/2}$–${}^2P_{1/2}$ fine-structure splittings of Al, Ga, In, and Tl are within 0.6% of experiment. For the super-heavy elements, a property of great interest is the ground-state configuration. Relativity changes the relative stability of the orbitals, leading frequently to ground states different from that of lighter elements in the same group of the periodic table. The chemistry of the element also changes for the same reason. Thus, eka-radon (element 118) is expected to have positive electron affinity, due to relativistic stabilization of its LUMO, the $8s$ orbital. Other examples will be shown, and the recent intermediate-Hamiltonian CC method, which extends the scope of Fock-space CC applicability and improves accuracy, will be discussed.