Computational Atomic Structures Toward Heavy Element Research

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Computational Atomic Structures Toward Heavy Element Research

by Sacha Schiffmann

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Abstract

We are interested in complex electronic structures of various atomic and ionics systems. We use an *ab initio* approach, the multiconfigurational Dirac-Hartree-Fock (MCDHF), to compute atomic structures and properties. We contribute in three main ways to the already existent literature: by developing and implementing original computer programs, by investigating possibilities of alternative computational methodologies and strategies, and finally by performing accurate atomic structure calculations to support other research fields, i.e., nuclear physics, astrophysics or experimental physics, through the theoretical estimation of relevant atomic data.

We raise questions about the choice of the optimal orbital basis by considering finite basis sets, MCDHF orbital bases and *natural-orbital* bases. We demonstrate the promising potential of the latter in the context of hyperfine structures and hope that others will find interest in pursuing our analysis. Ultimately, our work put forward some weaknesses of the traditional optimization strategy based on the layer-by-layer optimization strategy.

We also perform large-scale calculations to determine accurate atomic properties such as energy levels, hyperfine structures, isotope shifts, transition parameters, radiative lifetimes and Landé g factors. We show through the variety of atomic properties and atomic systems studied, the difficulty of describing, in the relativistic framework, the correlation between the spatial position of electrons due to their Coulomb repulsion.

This thesis is organized in two main parts. The first one is dedicated to the theoretical and computational backgrounds that are needed to understand the theoretical models and the interpretation of our results. The second part presents and summarizes our articles and manuscripts. They are separated in four groups, A, B, C, and D, around the themes of the atomic orbital bases, the applications to nuclear physics, the applications to astrophysics, and investigations of negative ions.

Key words

atoms, electron correlation, relativity, quantum mechanics, calculations, natural orbitals, isotope shifts, hyperfine structures, transition parameters, Landé g factors

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A doctoral thesis at a university in Sweden takes either the form of a single, cohesive research study (monograph) or a summary of research papers (compilation thesis), which the doctoral student has written alone or together with one or several other author(s).

In the latter case the thesis consists of two parts. An introductory text puts the research work into context and summarizes the main points of the papers. Then, the research publications themselves are reproduced, together with a description of the individual contributions of the authors. The research papers may either have been already published or are manuscripts at various stages (in press, submitted, or in draft).

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vous vous débattiez dans des problèmes si effroyablement compliqués
que même le jeune homme candide et optimiste
que vous étiez alors dut parfois,
comme ses camarades d'infortune,
maudire le jour où il avait eu l'idée saugrenue
de se mêler de physique atomique

Le principe - Jérôme Ferrari [Extrait]

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Popular summary

The world we live in is fascinating. Extraordinary is our ability to describe and communicate about any observed phenomenon with a single, universal language: mathematics. Throughout centuries humanity has developed models and theories to understand almost everything we witness. From the microscopic, infinitely small world to the macroscopic, infinitely large Universe, atoms are some of the most basic constituents we know. For long, atoms were believed to be unbreakable, so that by combining atoms we could explain the constitution of all matter. Nowadays, the existence of subatomic particles is established, revealing the compound nature of atoms. Each atom is composed of a small nucleus made of protons and neutrons and an electron cloud extending far out from the nucleus (the nucleus is 10 000 smaller than the atom).

We are interested in the properties of these atoms, and in particular we aim to understand and describe the collective behaviour of electrons. For that, we combine two of the most well-known theories of the 20th century: quantum mechanics and general relativity. The former is intricate with the infinitesimal nature of electrons and atoms, that results in a duality of characters –corpuscular and wave-like– for all matter, while the latter deals with high energy phenomenon and supplanted classical mechanics. Nevertheless, the dynamical systems formed by electrons and nuclei are extremely difficult to apprehend, and large computer resources are required to predict their properties.

Atoms and ions are immense sources of information thanks to the light they produce or absorb. Because of their quantum nature, electrons bound by the nucleus can only take discrete energy values, so that by jumping from one energy level to another, they emit or absorb light, revealing the corresponding atomic spectra. These atomic spectra are valuable to analyze the light incoming from e.g., distant stars, revealing the presence of certain elements and contributing to explain the origins of matter in the Universe. On a much smaller scale, these transitions help to characterize the atomic nuclei, their shapes, their sizes, etc.. In order to extract this useful information, theoretical calculations are required to predict accurately the electronic energy levels, transition energies and transition rates, including effects such as the interaction with the electromagnetic field of the nucleus or the influence of adding (or subtracting) neutrons in the nucleus (isotope dependence). The goals of this thesis are therefore to perform atomic structure calculations for a set of systems of interest and to contribute to the developments of computational methodologies and state-of-the-art atomic codes.

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