Electronic structure theory for high-precision spectroscopy, collisions, and attosecond processes

HEAD:

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GROUP MEMBERS:

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RESEARCH PROFILE:

Development of new tools of the electronic structure theory for an accurate description of high-resolution spectroscopic and collisional processes occurring in the ultracold regime, and also for the description of time-resolved electronic dynamics in the attosecond scale.

CURRENT RESEARCH ACTIVITIES:

New tools of the electronic structure theory for molecules in the ultracold regime

Molecules cooled to temperatures below T = 10^{-3} K allow for tackling questions touching upon the very fundamentals of quantum mechanics. They are promising candidates in novel applications, ranging from ultracold chemistry and precision measurements to quantum computing. Cold and ultracold molecules are thus opening up new and exciting areas of research in chemistry and physics due to their manifestly quantum nature. Understandably, to describe the processes occurring in the ultracold regime new tools of the electronic structure theory must be developed. In the course of the this research task we seek to:

- develop explicitly correlated coupled cluster methods for the ground state and the corresponding properties (dipole moments, electronic densities, etc.);
- apply Slater-type orbitals as a one-electron basis set and the explicitly correlated Kołos-Wolniewicz functions in the calculations for diatomic molecules;
- develop explicitly correlated electronic structure methods for excited states;
- generalise the expectation value formalism of the coupled cluster theory for the calculation of the coupled cluster linear and quadratic response functions at the explicitly correlated level;
- calculate coupling matrix elements (e.g., spin-orbit, nonadiabatic) between arbitrary excited states, as well as the transition moments and related quantities, based on the aforementioned newly developed methods;
- apply these newly developed methods to interpret current experiments in the ultracold regime.

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New tools of the electronic structure theory for molecules in the attosecond laser fields

Attoscience is a rapidly developing area of research with unparalleled application possibilities in fields of broadly understood chemistry, biology and physics. It focuses on processes occurring in extremely short timescale (1 as = 10^{-18} s) which allows for a direct study of the dynamics of electrons within atoms or molecules. Most of the attosecond processes involve irradiation of the sample (single atom, molecule, solid sample, gas sample etc.) with short but strong infrared impulses. This produces a variety of sample's responses, such as: (i) generation of high-order harmonics (HHG) by sample molecules, (ii) above-threshold ionization (ATI), accompanied by release of high-energetic free electrons, (iii) generation of multiply charged atomic and molecular ions, detection and analysis of which may be of value
in understanding the processes occurring within the electronic structure of molecules. However, without sufficiently developed theoretical description of attosecond processes these experiments may not fulfill their intended purposes. We are planning to create a range of advanced quantum chemistry methods for analysis and interpretation of gathered experimental data. These methods will be based on:

- numerical solutions of the time-dependent Schrödinger equation (TDSE) and it's non-linear version (TDNS) for one-dimensional atoms with one or two electrons, three-dimensional atoms with one or two electrons and one-dimensional molecules with one electron,
- the strong-field approximation (SFA) for one-dimensional atoms with one or two electrons, three-dimensional atoms with one or two electrons and one-dimensional molecules with one electron,
- classical Wigner methods for atoms, molecules and many-electron clusters,
- real-time time-dependent coupled cluster (TDCC) theory and density functional theory (TDDFT), which allow to obtain explicitly the time-dependent wavefuction and electron density for the many-electron systems;
- application of the electronic structure tools developed in our group for the time-independent case to an accurate description of time-dependent processes in the attosecond time scale.

SELECTED PUBLICATIONS:

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2. A.M. Tucholska, M. Lesiuk, R. Moszyński, Transition moments between excited electronic states from the Hermitian formulation of the coupled cluster quadratic response function, J. Chem. Phys. 146 (2017) 034108.
3. J.G. Balcerzak, M. Lesiuk, R. Moszyński, Calculation of Araki- Sucher correction for many-electron systems, Phys. Rev. A. 96 (2017) 052510.
4. M. McDonald, B.H. McGuyer, F. Apfelback, C.-H. Lee, I. Majewska, R. Moszyński, T. Zelevinsky, Photodissociation of ultracold diatomic strontium molecules with quantum state control, Nature. 535 (2016) 122.
5. T. Grining, M. Tomza, M. Lesiuk, M. Przybytek, M. Musiał, P. Massignan, M. Lewenstein, R. Moszyński, Many interacting fermions in a one-dimensional harmonic trap: a quantum-chemical treatment, New J. Phys. 17 (2015) 115001.
6. B.H. McGuyer, M. McDonald, G.Z. Iwata, M.G. Tarallo, W. Skomorowski, R. Moszyński, T. Zelevinsky, Precise study of asymptotic physics with subradiant ultracold molecules, Nature Physics. 11 (2015) 32.
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8. M. Lesiuk, R. Moszyński, Reexamination of the calculation of two-center, two-electron integrals over Slater-type orbitals. II. Neumann expansion of the exchange integrals, Phys. Rev. E. 90 (2014) 063319.
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