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D2.4 – Report on pre-release of an integrated platform with inter-operable Open Source flagship codes, including the I/O and, gradually, the QMCkl libraries

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Abbreviations

API	Application Programming Interface
CC	Coupled Cluster theory
CHAMP	Cornell-Holland Ab-initio Materials Package
CoE	Center of Excellence
DFT	Density Functional Theory
FCI	Full Configuration Interaction
HF	Hartree-Fock
MP	Møller–Plesset perturbation theory
MP2	second order Møller–Plesset perturbation theory
NECI	N-Electron Configuration Interaction
QMC	Quantum Monte Carlo
QMCkl	Quantum Monte Carlo Kernel Library
QP	Quantum Package
TREX	Targeting REal chemical accuracy at the eXascale
TREXIO	TREX Input/Output
VMC	Variational Monte Carlo
DMC	Diffusion Monte Carlo

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1 Introduction

The TREX project is in the field of high-accuracy quantum chemical and materials simulations with a special focus on Quantum Monte Carlo (QMC) approaches to the solution of the quantum many-body problem at the heart of atomistic physics, chemistry, and materials science. Importantly, due to their inherent parallelizability and high computational cost, QMC approaches, and thus TREX, are uniquely positioned to fully exploit the massive parallelism of the upcoming exascale supercomputer architectures.

The software developed in this project for improving code inter-operability and intefacing, will be referenced in the following as TREXIO. The library was released in the previous D2.2 deliverable, and is being continuously updated to facilitate its use and maximize its portability. This document will be the basis for the next D2.5 and the final D2.6 deliverable.

Let us remind the main idea of TREXIO that can be useful within and outside TREX CoEs. In a brute force approach, each developer, in order to interface its own code (e.g. TurboRVB) with any other code (see Fig.1, outer and inner shells), is forced to write a different interface for each of them, and this is a clearly inefficient waste of human resources. Instead with the TREXIO library, by means of highly developed and standardized input and output paradigms, only one interface is necessary: the one with the TREXIO library. This clearly represents an enormous simplification of the effort. Moreover, once all planned interfaces (green balls in Fig.1) will be available the visibility and the scientific appeal of the TREX codes, will be clearly enhanced.

In this report we will mainly focus on the pre-release of an integrated platform with inter-operable Open Source flagship codes, including the I/O. This library is important for the gradual integration of the QMCkl library (its status has been recently reported in the D3.2 deliverable) in the TREX flagship codes. All the technical aspects related to this important development will be dealt in the next D3 reports.

Scope

This report deals with the D2.4 deliverable, that is centered on the actions undertaken so far to develop TREXIO. The main purposes are to improve inter-operability both between the codes in TREX and to codes outside of TREX, to design a common data format and its implementation in an I/O library, as well as to build a well- documented set of reference test cases for the main functionalities of the codes.

Progress summary

- Created converters for data in file formats from codes outside of TREX (Gaussian, GAMESS and MOLCAS , already reported in D4.1 and, recently, PySCF, cclib and ORCA described in the last part of the next section). The library can be interfaced also (as reported in D4.1) with FCIDUMP and Molden formats.

- Use of TREXIO file format for increased inter-operability between the TREX codes Quantum Package (QP), CHAMP, QMC=Chem, TurboRVB, NECI, and GammCor.

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- Generated a preliminary reference set of test cases for the optimization/reproducibility of the main numerical tasks, within the same TREXIO input.

2 Interfaces to codes outside of TREX

The objective of the TREXIO library (available at github.com/TREX-CoE/trexio) is to facilitate inter-operability between codes in the field of quantum chemistry. In Fig.1 we report outside the dotted circle a list of codes that are pertinent to the project. The ones that are not explicitly marked are shown here only as possible codes to be included in the final release of the library, but should not be taken as a todo list for future developments of the library, because its extension is mainly driven by the users' requests. Indeed, in Fig.1 several external codes are clearly emphasized with explicit symbols, and represent the ones that have already been interfaced with TREXIO, or they are in an initial stage of development, and therefore remain in the "work in progress" status.

The TREXIO library is primarily focused on enabling the communication of data between the flagship codes of the TREX CoEs: NECI, GammCor, QP, QMC=Chem, CHAMP, and TurboRVB. Data elements such as, for example, the electronic wave function, reduced density matrices, and integrals are stored in a pre-defined format (see trex-coe.github.io/trexio/trex.html) in files. An Application Programming Interface (API) is provided to store and retrieve the data in TREXIO files. To maximize portability of the library, the source code is written in the C programming language, with bindings to Fortran and Python. To maximize uptake, the library is released under the permissive BSD 3-clause license.

The `trexio_tools` package (available at github.com/TREX-CoE/trexio_tools) is a collaborative effort to extend applicability and usage of the TREXIO library and corresponding file formats within and outside the TREX project. For example, the package contains a number of converters that can be used to transform the results of deterministic codes (see subsections below) into the TREXIO format, which can then be re-used for QMC calculations. It also contains utilities to test the self-consistency of TREXIO files. TREXIO is thus currently being actively developed as a tool to interface quantum chemistry codes both within and outside of the TREX CoEs. In the following subsections we summarize recent progress in this direction.

Gaussian and GAMESS

The TREXIO library has a binding for the Python language, and can easily be installed with the `pip` Python package manager. The `resultsFile` Python package, authored by Anthony Scemama (CNRS-Toulouse) and available at gitlab.com/scemama/resultsFile, can fetch all the data required for QMC calculations from output files produced with the Gaussian [1] and the GAMESS [2] programs, and historically is the package used to convert GAMESS/Gaussian output files to input data for QP. As part of the `trexio_tools` package, we have written a converter that uses `resultsFile` to read the output of GAMESS/Gaussian, and that writes out the wave function into a TREXIO file. For this converter to read a Gaussian output file, the keywords `GFPRINT` and `pop=Full` need to be specified in the Gaussian input since Gaussian does not print by default all the virtual orbitals and the basis set parameters.

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MOLCAS

The Python API of the TREXIO library enables many possibilities to pre-process the quantities required for a QMC calculation and convert them into different file formats via Python scripts. The easy accessibility of a Python package manager helps in using these scripts from the command line. Similar to the `trexio_tools` package (which handles conversion of GAMESS/Gaussian output into TREXIO formats, see above), we developed a `qc2champ` Python package. With this package, the output of a MOLCAS [3] or openMOLCAS [4] calculation can be parsed and all the required input datafiles for a CHAMP calculation or the general TREXIO file format can be generated. The package is available on PyPI and can be easily installed with the `pip` Python package manager via the command `pip install qc2champ`. The MOLCAS is a stable version of openMOLCAS, that is the developer branch, containing the most recent updates. Obviously our converter works also for OpenMOLCAS, as long as the most recent updates do not change the data format, as it is the case at present.

PySCF

pySCF (see https://pyscf.org) is an open source code written in Python, that contains the most advanced quantum chemistry algorithms for ground state and electronic excitations of molecules, as well as for periodic systems and bulk properties. pySCF implements various quantum chemistry methods such as Hartree-Fock (HF), Density Functional Theory (DFT), Møller–Plesset perturbation theory (MP), and Coupled Cluster theory (CC). Since pySCF is interfaced with Libxc (see https://tddft.org/programs/libxc), a user can employ various exchange-correlation and kinetic energy functionals for DFT calculations. The TREXIO library has been recently extended to deal with this important quantum chemistry package so that all quantum Monte Carlo codes, CHAMP, TurboRVB and QMC=CHEM can interface now with this important software, and take advantage of relevant tools such as MP, expansion in natural orbitals, couple cluster theory that are not commonly available in open source distributions.

ORCA

ORCA is an open-source ab initio quantum chemistry program package that contains modern electronic structure methods including density functional theory, many-body perturbation, coupled cluster, multireference methods, and semi-empirical quantum chemistry methods. Its main field of application is larger molecules, transition metal complexes, and their spectroscopic properties.

Due to the user-friendly style, ORCA is considered to be a helpful tool not only for computational chemists, but also for chemists, physicists and biologists that are interested in developing the full information content of their experimental data with help of calculations. Work is in progress to develop an interface capable to translate the ORCA outputs in TREXIO files, that can be later processed by any flagship code of TREX.

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cclib

cclib is an open source library, written in Python, for parsing and interpreting the results of computational chemistry packages. The goals of cclib are centered around the reuse of data obtained from these programs and contained in output files, specifically:

- extract (parse) data from the output files generated by multiple programs.
- provide a consistent interface to the results of computational chemistry calculations, particularly those results that are useful for algorithms or visualization.
- facilitate the implementation of algorithms that are not specific to a particular computational chemistry package to maximize interoperability with other open source computational chemistry and cheminformatic software libraries.

3 Data formats supported in TREXIO

TREXIO is at present compatible with other formats such as:

FCIDUMP

The FCIDUMP format was initially the format used by the Full Configuration Interaction (FCI) code of Hans-Joachim Werner, which was then included in Molpro [5]. Although this format has all the disadvantages of text-based formats for large files (file size, and slow to parse), this format has become widely adopted in the community by codes that only require one- and two-body integrals as an input, because of its simplicity. Hence, we have added a converter between TREXIO and the FCIDUMP format to increase adoption of the TREXIO file format by external codes.

Molden

Molden [6] is a code that visualizes a three-dimensional rendering of a molecule, its molecular orbitals, and electron density. It's so-called Molden format (see www3.cmbi.umcn.nl/molden/molden_format.html) has been widely adopted by quantum chemistry codes such as ADF, MOLCAS, Molpro, and Dalton to store molecular orbitals. To increase the compatibility of the TREX software we have also written a converter from the TREXIO file format to the Molden file format.

4 Interfaces between TREX codes

The inter-operability between the TREX flagship codes relies on the TREXIO library (see Figure 1). In the following subsections, we summarize recent progress in this direction.

Quantum Package

Quantum Package (QP) [7] is a deterministic quantum chemistry code which can be used as an entry point for all TREX calculations, as it can compute trial wave functions using standard methods of quantum chemistry including Hartree-Fock, density functional theory, configuration interaction, and

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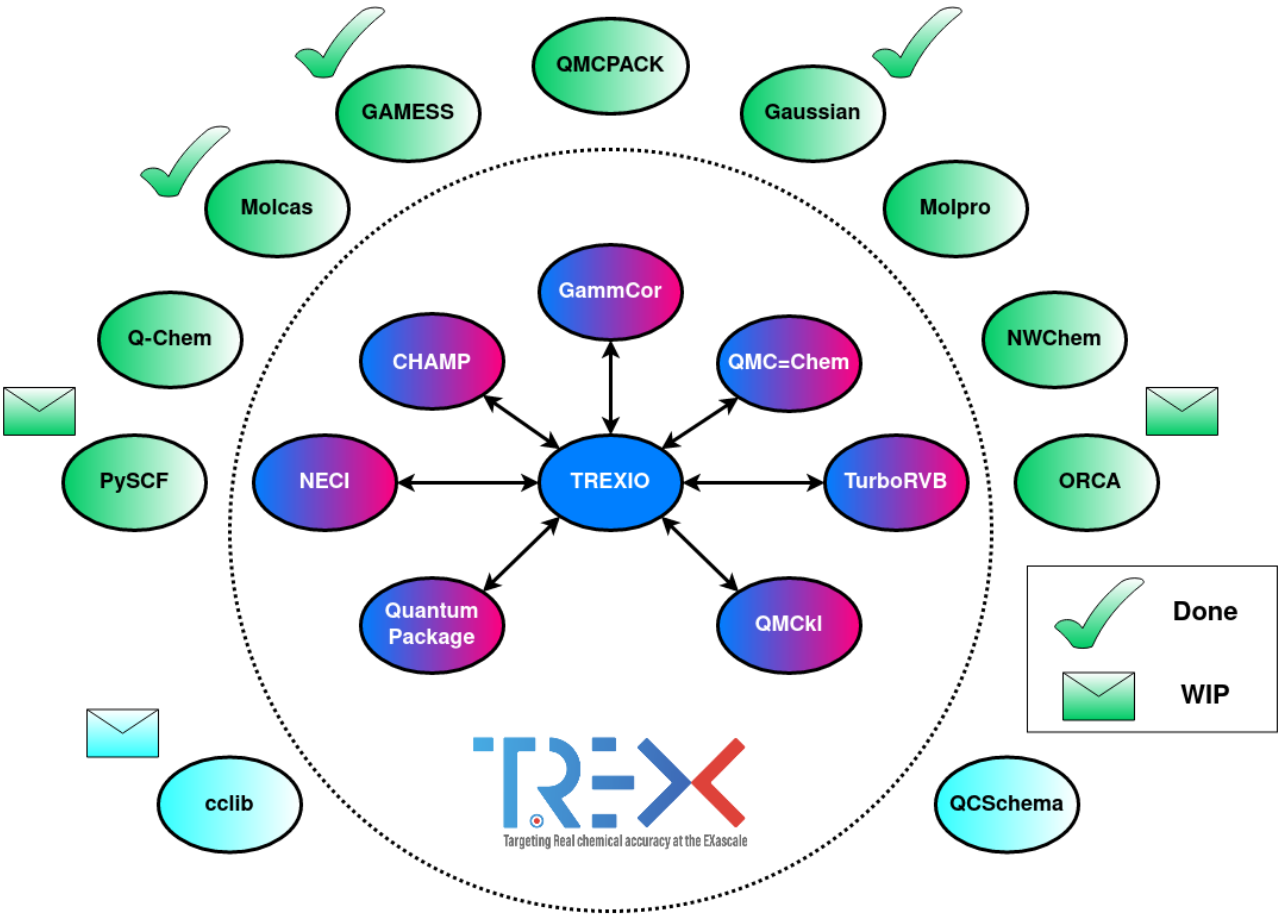

Figure 1: *Schematic diagram of inter-operability of quantum chemistry codes* (ellipses) within the TREX CoEs (inside the dotted circle) and outside TREX. Among these codes, the marked ones support a fully working interface with TREXIO, while the ones surrounded by an envelope are "work in progress" and have already been used in some applications.

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others. It has hence been the first code that was fully interfaced with TREXIO, with support for atomic coordinates, basis sets, effective core potentials, molecular orbitals, one- and two-electron integrals, and one- and two-body reduced density matrices, as well as reading data from TREXIO files, allowing to run workflows initiated from QP.

CHAMP

Cornell-Holland Ab-initio Materials Package (CHAMP) [8] is a suite of programs for QMC electronic structure calculations of atomic and molecular systems. It enables efficient wave function optimization for ground and excited states, a compact formulation for a fast evaluation of multi-determinant expansions and their derivatives, as well as multi-scale schemes to perform QMC calculations in classical point charge models, polarizable continuum models, and polarizable force fields.

QP and CHAMP codes were historically interfaced by a series of involved scripts that required extensive human intervention. CHAMP has now been interfaced with TREXIO so that all data present in a TREXIO file can be read by CHAMP. To date, the only missing information in TREXIO needed for a CHAMP calculation are the coefficients of the determinant expansion. As a work-around, a simple plugin was developed in QP to write out this information in CHAMP format, to be used until this information becomes available in TREXIO itself.

QMC=Chem

QMC=Chem [9] and QP are both written by the same developers, and have been interoperable since the creation of QP. We are now working on using the TREXIO format as a native format for the storage of the wavefunction in both codes.

QMC=Chem had implemented only a minimal Jastrow factor together with a basic optimization algorithm. However, the form of the Jastrow factor used in CHAMP has been implemented in TREX Quantum Monte Carlo Kernel Library (QMCkl). By linking with QMCkl, the Jastrow factor of CHAMP is now accessible to QMC=Chem with minimal effort. As both QMC=Chem and CHAMP can use trial wave functions generated from QP, the effort of writing a smart optimizer for the Jastrow factor can be avoided by using CHAMP to optimize the Jastrow parameters and re-use them in QMC=Chem.

TurboRVB

TurboRVB [10] (where RVB stands for Resonating Valence Bond) is a computational package for ab initio QMC simulations of both molecular and bulk electronic systems. The code implements two types of well established QMC algorithms: Variational Monte Carlo (VMC) and Diffusion Monte Carlo (DMC) in its robust and efficient lattice regularized variant. TurboRVB has been successfully interfaced with TREXIO. However, TurboRVB works internally using atomic basis sets in spherical coordinates. As QP works only with Cartesian coordinates, TurboRVB cannot yet be used with TREXIO files produced with QP. Recently a converter from spherical to Cartesian coordinates has been developed and TurboRVB outputs can be read and elaborated by all the quantum chemistry packages that work in cartesian coordinates. In the near future the corresponding converter from

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Cartesian to spherical will enable TurboRVB to read data without restriction from any packages interfacing with TREXIO.

NECI

N-Electron Configuration Interaction (NECI) [11] is a FCI QMC implementation, based on a stochastic application of the Hamiltonian matrix on a sparse sampling of the wave function. Previously, as NECI can use FCIDUMP files, QP and NECI would communicate by generating a FCIDUMP file directly from QP. It is now possible to convert a TREXIO file into an FCIDUMP file. Thus, all codes that can read the one- and two-electron integrals from FCIDUMP format are now interfaced with TREXIO. At the same time, this is another communication channel from QP to NECI, using the TREXIO file in the middle.

GammCor

GammCor [12] performs calculations of interaction energies using multi-reference symmetry-adapted perturbation theory and the adiabatic connection correlation energy for ground- and excited-state multireference wave functions. GammCor uses one- and two-electron integrals as an input, together with one- and two-body reduced density matrices. The reduced density matrices can be computed by both NECI and QP. By now, QP is able to store all the required integrals as well as the density matrices in TREXIO file format. We are now working on reading these quantities into GammCor from the TREXIO files, and writing the density matrices in the TREXIO file from NECI.

5 First release of a set of reference cases for the optimization/reproducibility of the main numerical tasks, within the same TREXIO input.

All TREX codes have a rather complex structure and require advanced programming skills for their reliable implementations. Moreover, statistical methods, such as the ones based on the so called quantum Monte Carlo method, have the difficulty to reproduce results at machine precision. The statistical errors may be rather large for large and complex systems. For this reason we have focused on a very simple and well studied system consisting of a few atoms:

1. The Hartree-Fock of the benzene molecule consisting of an hexagonal ring made of six Carbon and six hydrogen where the Carbon-Carbon (Hydrogen-Hydrogen) distance is set to 2.4 a.u. (3.2 a.u.). The Hartree-Fock solution can be evaluated exactly on a given basis and with a given psuedopotential with well established quantum-chemistry packages. Here, the ccECP pseudopotentials and the corresponding contracted cc-pVDZ basis sets were used. The real spherical notation is employed for constructing the atomic orbitals. The obtained total energies by different quantum-chemistry packages are summarized in Table 1. This quantity has been computed by several TREX-codes by means of quantum Monte Carlo. Though a statistical method is not necessary for such a simple wavefunction, the present setup is clearly useful to

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Table 1: Comparisons of HF calculations

Software	Total Energy (Ha)
Gaussian	-36.5055
PySCF	-36.5055
Quantum Package	-36.5055

test the TREXIO library and the correctness of the TREX codes in the interface with this library. Indeed, in this test, CHAMP, QMC=Chem and TurboRVB, have used the same TREXIO input and the statistical accuracy has been pushed to a limit that does not leave doubts on the consistency of the results, because, as shown in Table 2, all results are mutually consistent within twice the error bars, and in turn with the exact energy reference of the previous table.

Table 2: Comparisons of the HF energy evaluation by using some of the TREX-flagship codes, that implement the quantum Monte Carlo method. The outcome result is therefore affected by a statistical error in the last digit and reported between brackets.

Software	Total Energy	Variance
CHAMP	36.5057(1)	7.004(7)
QMC=Chem	-36.5060(4)	6.999(5)
TurboRVB	-36.5053(1)	7.017(6)

2. In order to check also the simplest correlated calculation, when no reference outside the QMC community is available, we have considered an extension of the previous Hartree-Fock wave-function, with a standard correlation term[13], characterizing the mutual repulsion of electrons when placed at a distance r:

$$u(r) = 1/2\frac{r}{1+br}. \quad (1)$$

With the parameter b kept fixed to the reference value $b = 4$, all codes show consistent results within statistical errors, as far as the energy is concerned (see Table 3). As far as the variance of the energy, a small discrepancy is observed between TurboRVB and QMC=Chem, within the DLA approximation. In this respect one has to consider that the variance may depend much more on the approximations used in the various codes for the pseudopotential integration. For instance TurboRVB uses a finite integration mesh, whereas QMC=Chem uses single precision for evaluating atomic and molecular orbitals.

Finally we have checked the performances (in the total time spent) and the interoperability of TREXIO for reading/writing the four index table, that is necessary to express the realistic Hamiltonian in a localized basis and is in turn important for several applications of GammCor and NECI. In particular, incorporating TREXIO in GammCor and Quantum Package - two flagship TREX codes - has

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Table 3: Comparisons of VMC calculations with the simple Jastrow. The computations labeled with DLA are obtained without considering the Jastrow in the non local part of the pseudopotential. The statistical error affecting the last digit is reported between brackets.

Software	Total Energy	Variance
CHAMP	-36.6415(2)	4.5177(5)
QMC=Chem (DLA)	-36.6410(4)	4.479(1)
TurboRVB	-36.6415(1)	4.5183(4)
TurboRVB (DLA)	-36.6413(1)	4.4956(4)

opened a way for efficient exchange of data between the codes. Two types of objects are transferred via TREXIO to GammCor,1) the four-index tables: two-electron integrals and two-particle reduced density matrices and 2) two-index tables: one-electron Hamiltonian and one-particle reduced density matrices. They are generated in Quantum Package from wavefunction calculations and written in TREXIO format. In a second step they are read in GammCor and used for the computation of molecular interaction calculation and analysis. In an upcoming release we plan to enhance the synergy of the codes by reducing dimensionality of the four-index tables managed by TREXIO, by replacing them with three-index objects obtained using Cholesky decomposition techniques, applicable to positive-definite matrices. As far as the NECI package is concerned, the developers are currently working on a TREXIO compatible library called TCHINT, which generates 4-index and 6-index integral files, suitable for the standard Schrödinger Hamiltonian, as well as Jastrow-factorised similarity-transformed (or Transcorrelated) Hamiltonians, where the Jastrow factors have been obtained from a variational Monte Carlo code. This will allow NECI to run quantum chemical calculations based on Jastrow factors obtained from other TREX VMC codes, and also allows other quantum-chemistry-based TREX codes (such as Quantum Package) to run similar calculations based on the transcorrelared integrals obtained from the TCHINT library.

6 Conclusions and perspectives

The present pre-release of the TREXIO library has been a success: it has allowed the benchmark of several TREX-flagship codes and, even more importantly, will enable users to perform new calculations thanks to the interoperability of codes within and outside TREX.

Within TREX, for example, QP, CHAMP, and GammCor can be now used "together" for very accurate multi-reference calculations of weak interactions by exchanging inputs and outputs by means of TREXIO. Due to interoperability with codes outside TREX, new calculations are now possible as for instance with the use of natural orbitals determined within second order Møller–Plesset perturbation theory (MP2) with external codes such as GAMESS/Gaussian/PySCF. In this way, one can accelerate the convergence of multi-configuration expansions or construct better wavefunctions for single-configuration QMC calculations in large systems. The latter wave functions can be also used for better initialization in the optimization of variational wavefunctions such as Antisymmetrized

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Geminal Product (AGP) or Pfaffian wavefunction that have an intrinsic but very entangled correlation structure. This entanglement is usually very difficult to exploit without external inputs.

Thanks to TREXIO, we have here provided a first set of test cases where the TREXIO library is used for sharing the same input among different TREX-codes. This first experiment was useful though limited to the case of a single benzene molecule and a small basis set. We plan to extend this test set to include more complex molecules (e.g. transition metals) and larger basis sets (up to quintuple zeta). Moreover, since it is difficult to check codes with outputs affected by statistical noise, in the next release of the library, we will select a number of electronic configurations where a deterministic answer will be provided. Indeed, the so-called local energy corresponding to a given electronic configuration, a key computational task for any QMC code, can be decomposed in its various components:

1. the bare kinetic energy,
2. the contribution of the local part of the pseudo,
3. its non local part,
4. the Coulomb electron-ion attractive term,
5. the Coulomb electron-electron repulsive term.

With the next release of TREXIO, it will be possible to read a given electronic configuration and all flagship codes computing the local energy will have to produce consistent results within machine precision (points no. 1,2,4,5 above) or the tolerance accuracy in the evaluation of the pseudo integration (point n. 3 above).

In summary, the development of the TREXIO library is proceeding extremely well and at high speed (even beyond our original expectations). We believe that, when this library will be completed, it will represent a fundamental tool for targeting high accuracy in quantum simulations with high performance computers.

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