3Ba05

UTChem - A Program for *ab initio* Quantum Chemistry

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Present molecular quantum theory is highly sophisticated, and has evolved dramatically. Software forms a basis for computational chemistry. However, it is not an easy task for an individual/group to develop a comprehensive new program package in *ab initio* quantum chemistry from scratch. Several years ago, we decided to accept this challenge. In view of the availability of such good programs as Gaussian, Gamess, Molcas, NWChem, etc., one may question the relevance of a new program package. We have three arguments for our project. (1) First, we believe that healthy competition is very important in science. (2) Second, we can have a good harvest by doing research using other programs, but it is an abortive flower. We could not make a true breakthrough if we were circumscribed by current software limitations. (3) Third, in spite of the excellent performance of other programs, there are important and powerful methods that others cannot yet handle. We have developed new methodologies in quantum chemistry, particularly the multireference-based perturbation theory for describing chemical reactions and excited states, relativistic molecular theory to treat heavy elements, parameter-free (less) exchange and correlation functionals in DFT, highly efficient algorithms for calculating molecular integrals over generally contracted Gaussians, etc. UTChem is a research product of our work to develop new and better theoretical methods in quantum chemistry. Most of the codes have been developed recently by Hirao's group at the University of Tokyo. The basic philosophy behind UTChem is to develop methods that allow an accurate and efficient computational chemistry of electronic structure problems for molecular systems in both the ground and excited states. UTChem also contains codes for well-developed methods such as MPn, CI, CC, etc., which are standard in most quantum chemistry programs. We are aiming ultimately at better performance than other programs. UTChem will soon be ready for distribution. Here you will be able to see the features of UTChem. UTChem contains a large number of improvements and some interesting new features, which others cannot match.

Synthetic design and conception for implementation of UTChem

A packaging of UTChem got started in the situation that researchers and students in Hirao laboratory had accomplished their implementations individually on independent developmental environments. The accomplishments are covering wide range of capabilities enough to do a sequence of *ab initio* calculations without using other extant distributions as follows,

1) multireference perturbation method (MRMP, MCQDPT),
2) multiconfigurational self-consistent field method (QCAS, GMC, CASVB),
3) relativistic 1/2-component method (RESC, DK3),
4) relativistic 4-component method (DHF, DKS),
5) one- and two-electron integrals,
6) direct and conventional self-consistent field method,
7) density functional theory (OP, PFREE),
8) dynamics and simulation involving QM/MM,
9) response theory for Hartree-Fock and density functional theory.

Our synthetic design and conception are basically coming from a practical necessity on how smooth and stressless compilation of the above existing program codes can be carried out. The fundamental structure we desire is that UTChem should be a program suite which enables developer groups to develop separately executable programs of their owns individually with no consolidation into one binary. This approach is based on the development philosophy that programmer groups can program as freely as possible with no disturbance of serious problems and fatal bugs of the damaged codes the other programmer groups have committed, and no intruder of the other developer groups into their own pretty codes and motivations.

Technically, through a sequence of calculations using separated binaries, what the individual program binaries share with each other is data files compliant with the file formats the developers have specified. UTChem itself just governs a number of rules of the file interfaces, (i.e., no subroutine interfaces, no program rules, ...), which are an exclusive way to connect the individual executable binaries. This design is simple and widely used in other program systems, meanwhile a method to control several program binaries has to be implemented so as to distribute them as a reliable and usable program suite. In UTChem, Python language is used to provide a high-level and flexible script to control and manage the calculation flows consisted of separately executed programs. We believe that this implementation also provides an accessibility, in which various developers also easily contribute their own program to UTChem.

As to practical calculation flows, figure 1 depicts a flowchart in UTChem. After the frontend program read a given input file, required one- and two-electron AO integrals are calculated. HF/KS-SCF as well as TD-HF/DFT for excitation energies and Local MP2 are carried out with 2e AO integrals directly-driven or restored from disk. The single-reference electron correlation theories such as configuration interaction theory (CISD, CISDT, CISDTQ), coupled-cluster theory (CCD, LCCD, CCSD, LCCSD, QCISD, CCSDT, CCSDTQ), and Møller-Plesset perturbation theory (MP2, MP3, MP4) achieved by tensor contraction engine (TCE) are following the integral transformation. Multiconfigurational calculations including MCSCF and perturbation (MRMP and MCQDPT) method are connected with AO integral part and HF-SCF part. After the above single-point energy calculations, if necessary and executable, the derivative energies with respect to the nuclear coordinates are derived. The gradients are used in the geometry optimization code and dynamics simulation including QM/MM.

Figure 1. Schematic calculation flowchart in UTChem