J|A|C|S
COMPUTER SOFTWARE REVIEWS

MOLCAS 7.2. MOLCAS, Department of Theoretical Chemistry Lund University, Chemical Center, P.O. Box 124, S-221 00 Lund, Sweden. http://www.teokem.lu.se/molcas/. See Web site for pricing information.

MOLCAS is a sophisticated and comprehensive quantum chemistry software package for the study of electronic and structural properties of molecular systems. It is not primarily a commercial product but a program developed by scientists at Lund University (Sweden) and others in the MOLCAS network around the world. Currently most users are in academia, although there are a limited number of industrial users. Available in source code, it can be configured for Unix, Linux, MS Windows, and Mac OS. Several 3.06 GHz iMacs with 4 GB RAM running Mac OS X 10.5.5 were used for this evaluation. Mac users should be aware that MOLCAS 7.2 may not be as easy to configure for Mac OS as for Windows or Linux.

Although MOLCAS can perform all the standard calculations on single-determinant reference states common to most quantum mechanics packages (HF, DFT, MP2, CC, and CPT), the key features of MOLCAS are well-developed codes for multiconfigurational SCF calculations (MCSCF) at the Complete Active Space (CASSCF) and more restricted RASSCF levels. Such calculations are especially suitable for treatment of highly degenerate states occurring in excited states, transition states, diradicals, and heavy metal systems, for example. In addition, dynamic electron correlation effects can be treated using CASPT2, the MOLCAS version of multireference perturbation theory that remains under constant development as one of the more successful such treatments. Although development of an analytical gradient code for CASPT2 remains incomplete, a numerical gradient code became available with Version 6 that allows for optimization of geometries at the CASPT2 level and is applicable in all areas where CASPT2 energies can be computed.

Three sets of generally contracted basis sets have also been designed especially for MOLCAS to give a balanced description of the atoms in ground, excited, and ionized states. Also introduced in Version 7 is a Cholesky decomposition of the two-electron integrals that has substantially increased the size of systems and basis sets that may be treated at all levels of theory.

The authors correctly note in the User's Manual that MOLCAS is not a "black box" method and that users need to have familiarity with various quantum mechanical methods as well as the ability to analyze results critically. However, there are many resources available to help users develop their skills. In addition to serving as a guide to installation, programs, and the MOLCAS environment, the User's Manual (www.teokem.lu.se/molcas/documentation/manual/manual.pdf) includes both examples and tutorials, the latter being broken down into problem- and program-based ones. There is a bulletin board and a mailing list (www.teokem.lu.se/molcas/wwwboard) where help can be obtained from other users and developers alike. The MOLCAS group also conducts workshops. Although the first four were held in Europe, the fifth one will be held in Minneapolis, Minnesota, USA on May 4−8, 2009. I attended one last year, which proved to be a highly valuable experience, albeit a rather intensive one.

In addition, there have been major improvements in the user-friendliness of MOLCAS with Version 7.2. These include improvements to the input language, including new commands (EMIL) for file handling, and an enhanced platform independent GUI that includes both a grid viewer for MOs, etc., and a graphical input generator (MING). The latter will likely be helpful for the novice user in learning how to prepare input files. The software remains modular in design, employing a number of interdependent programs or modules identified with an "&" prefix (see below). Several of my undergraduate research students have become reasonably proficient in using MOLCAS.

In this evaluation, emphasis was placed on performing CASPT2 calculations, both single-point and geometry optimizations, beginning with structures whose coordinates were taken from previous CASSCF calculations performed using Gaussian03. With previous versions of MOLCAS, obtaining CASPT2 energies this way necessitated running an HF SCF calculation (&SCF) on the structure obtained after computing the one- and two-electron integrals (&SEWARD), after which the active-space orbitals always had to be altered before proceeding to the CASSCF (&RASSCF) and subsequent CASPT2 calculations (&CASPT2). With Version 7.2, I found that the SCF calculation step could be omitted with the resulting CASSCF active-space orbitals remaining unaltered, presumably due to the implementation of a "GuessOrb" facility in &SEWARD. This represents a significant advantage over Version 6 in terms of convenience and time. I also found it straightforward to obtain CASPT2-optimized minimum structures; however locating optimized transition structures in general proved more problematical than with Gaussian03. Perhaps greater facility with the documented procedures for running transition state searches will come with more practice. As CASPT2 optimizations are costly, it is strongly recommended that they be run in parallel in a multiprocessor environment. The parallel version of MOLCAS has been tested under Linux and Fujitsu Solaris, but not under Mac OS X.

MOLCAS 7.2 is recommended for scientists with a moderate to excellent understanding of MCSCF computational methods, either as a stand-alone package or as a supplement to other computational software programs.

James A. Duncan,
Lewis & Clark College

JA900300H

10.1021/ja900300h

10.1021/ja900300h CCC: $40.75