研究工作詳述

簡介

圓二色光譜(circular dichroism, CD)是分析有機化合物絕對立體化學的有效方法。近年來由於電腦軟硬體成長快速，自 2000 年起在國際期刊逐漸出現以 calculated ECD 為判斷化合物立體化學結構的文章，近年更成為重要參考依據。量子化學軟體 Gaussian 09 的版本在 vibronic computations 已包括程式碼最佳化的 ECD 運算功能，在搭配 time-dependent (TD)的 time-dependent density functional theory (TDDFT)計算中。

問題描述

在個人電腦中進行 Gaussian 運算，需耗數天至數十天時間。如在高效能運算服務的設備下進行運算，時間將可縮短數倍到數十倍的時間，對提升研究速度的幫助極大。

擬採用方法

STEP 1: DEFINE THE BOUNDRIES

The calculation will be started by defining how much time, energy and CPU are available for the study.

STEP 2: SET UP THE COMPUTER

Turn off all the other services (if you're not sure, skip this). Make sure your computer does not go into sleeping or hibernating mode automatically after some period of time, this might kill a G09 job.

STEP 3: DEFINE A GOOD WAY TO NAME THE JOBS

This notation puts the name of the molecule followed by the MM method that was used initially to optimize the geometry, followed by the theoretical model, followed by the basis set and finally by the type of calculation performed.

STEP 4: DEFINE A PARENT MOLECULE

Starting with a parent molecule will aid us to choose the right theoretical model and basis set. Run several calculations using the same theoretical model, but with increasingly better basis sets, compare with literature values. Then restart with a different model. This will show you which combination outputs the smallest error, and therefore suites the best for this case.

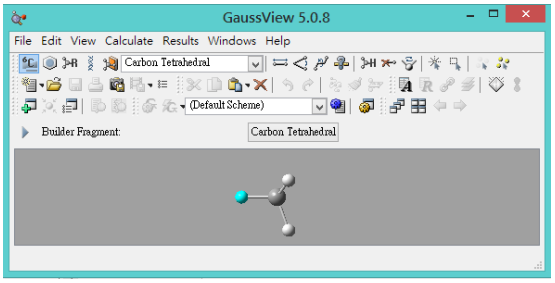
STEP 5: SUCCESFULL SEQUENCE OF CALCULATIONS

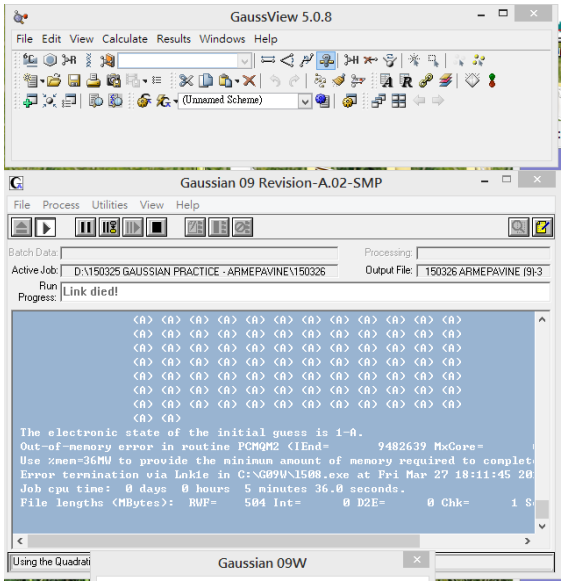
The calculation will be performed by the following sequence of calculations for any molecule:

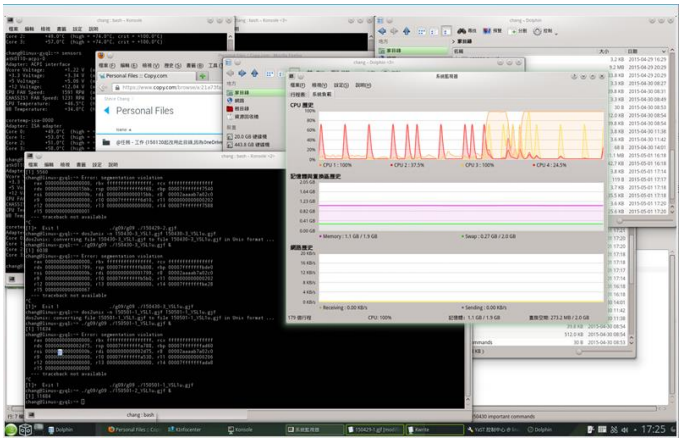
1. Geometry Opt in Avogadro using MM (UFF or MMFF).
2. Geometry Opt and Freq calculation in G09 .
3. Check for imaginary frequencies (none for ground state, 1 for TS, etc).

4. If frequencies are alright, continue, otherwise restart the geometry opt.
5. Compute the ECD spectrum.
6. Re-opt and re-compute frequencies in solvent (if needed).
7. Re-compute the ECD property in solvent (if needed).

Some screeshot of the Gaussian calculations







預期成效

The absolute configurations of 1, 2, and 4 were supported by quantum chemical calculations of the ECD spectra. The structure of (2S,3R,4S,6R,7R)-1 was optimized with the density functional theory (DFT) using the hybrid functional B3LY 9 and the correlation-corrected basis set aug-cc-pVDZ. 10 The ECD spectrum was calculated with the time-dependent density functional theory (TDDFT) 12 using the same functional, B3LYP, 9 the basis set aug-cc-pVDZ, 10 and the C-PCM solvent model 11 for acetonitrile. The quantum chemical calculations were carried out with Gaussian 09. 13 The calculated ECD

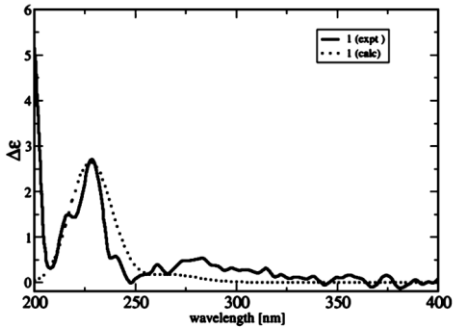


Figure 4. Experimental and calculated ECD spectra of **1**.