Tata Institute of Fundamental Research
Survey No. 36/P, Gopanpally Village, Serilingampally, Ranga Reddy Dist., Hyderabad - 500107

Internal Webinar

Rapid and Accurate Composite ab initio Models for High-throughput Computation of Molecular Thermochemistry across Chemical Space

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The MolD is computational chemistry Big Data analytics platform (https://moldis.tifrh.res.in/) aims at developing various domain specific molecular and materials datasets. For molecular datasets relevant to organic- and bio-chemical applications, determination of thermochemical and kinetic properties are vital. Although, sophisticated quantum chemical methods can predict the molecular energies to fine precision, they also incur significant computational cost – making them impractical for large systems. This led to the rise of quantum chemistry composite methods which can estimate thermochemical properties accurately within reasonable computer time. The Gaussian-n (G_n) theories, belong to a class of composite methods, which can predict the thermochemical properties precisely, but at the same time with affordable computational cost. The G4 method is so far the most accurate variant amongst the G_n theories and is able to precisely estimate the aforementioned properties within sub-kcal region. The more economical variant of G4 is G4MP2, which reduces the computational cost while making some compromise with the accuracy.

The major focus of this study is on enthalpy of formation, but at the same time, we have also explored other thermochemical properties like heat-of-formation, ionization energy, electron affinity, proton affinity, non-bonding interaction energy and pKa. Critical evaluation of the additivity assumption in the composite procedure G4MP2 is done, in-order to propose modifications to this scheme which can further reduce the computational cost without compromising accuracy and transferability across various chemical datasets. The proposed strategies are based on the theories of resolution-of-identity (RI) expansion of two-electron Coulomb integrals and local coupled-cluster method. The RI approximation replaces the four-index integrals by three and two-index quantities, via the use of auxiliary basis set and reduces the time in computation of largescale integral transformations, prevalent in the theoretical calculations. The local coupled-cluster approximation restricts the correlation space and scales significantly with increasing no. of atoms. Preliminary results indicate that this new method forecasts molecular energetics with in about 1 kcal/mol accuracy compared to pruned experimental datasets and enables high-throughput computation of molecular thermochemistry.

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