DEVELOPMENT OF HIGH-SPEED AB INITIO CCSD(T) LEVEL NEURAL NETWORK POTENTIAL ENERGY SURFACES FOR DIFFUSION MONTE CARLO

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Diffusion Monte Carlo (DMC) is a general statistical method that is capable of providing an accurate ground-state solution to the molecular Schrodinger equation of the system of interest. The approach is particularly well suited for systems like water clusters and CH_5^+ that undergo large amplitude vibrational motions, providing a way to gain insights into their vibrational and rotational spectra that are difficult to achieve by other methods. The ability to perform DMC simulations is predicated by the availability of a fast and reliable Potential Energy Surface(PES), as billions of structures with energies up to ten times the zero point energy will be evaluated in a typical DMC simulation. Such strenuous demands for speed, accuracy and extrapolability to high-energy regions of the potential pose major challenges to most current PES developed by conventional methods.

To address these issues, we have developed a Neural Network(NN) architecture and training protocol to generate CCSD(T) level NN-PES specifically to meet all the demands of DMC. We validated this approach with CH_5^+ and $(H_2O)_2$, and applied it to protonated ethylene ($C_2H_5^+$). This proposed NN-PES is trained solely with *ab initio* data, and is versatile, so it can be applied to any small-to-medium-sized systems. Powered by the robust parallel computing ability of Graphics Processing Units (GPUs), this approach can be used to evaluate the energy of a single geometry with a microsecond. Its architecture also ensures remarkable extrapolability and no unphysical energy predictions (e.g. 'holes' in the potential) even in high energy regions of the potential where training data are extremely scarce. In this talk we will focus on the procedures taken to develop such NN-PES, and in the accompanying talk, we will share the results of DMC studies of $C_2H_5^+$ that use a NN-PES, which has been developed using this approach.