Composite Gaussian processes for probabilistic PES prediction

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The assignment of spectral lines in both the visible region and the UV region has important aplications in the study of astral objects such as stars and exoplanets, or, closer to us, in the study of the molecular composition of the terrestrial atmosphere. The correct assignment of spectra cannot rely solely on experimental results since those are not always available making theoretical predictions necessary.
To this end, we extended the use of Gaussian processes in PES prediction[1] to the use of composite machine learning Gaussian *regression* processes (c-GP) trained at different levels of theory, with different training sets, is explored. The final energy is a sum of composite probabilistic prediction corresponding to dense training sets at low levels of theory and sparse training sets on computationally expensive deterministic and stochastic[2,3] methods.
The study of the H_3^+ molecular ground singlet and triplet states and the performance of a prediction based on the HF surface with a correction based on the difference between the deterministic CCSD (equivalent to the FCI energy for H_3^+) energy and the HF energy is presented.
The prediction of both vibrational and rotational energy levels using the DVR3D[4] method is then applied to the obtained probabilistic PES for the ground singlet state.

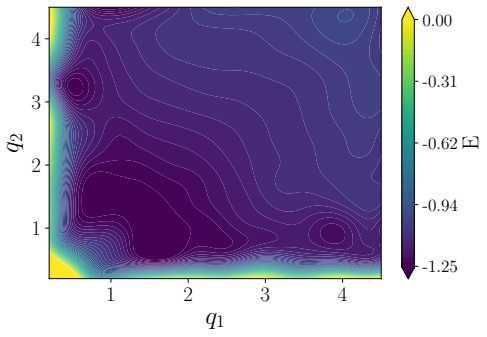

Figure 1: c-GP prediction for H_3^+

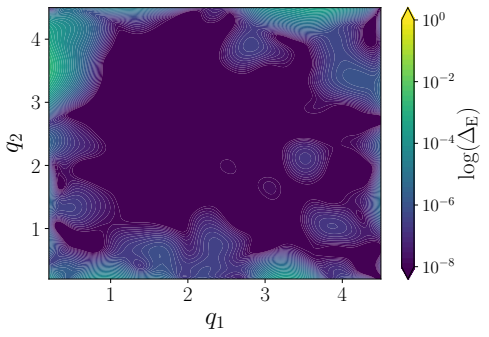

Figure 2: c-GP 95% confidence interval

References

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