

# In Pursuit of High-Accuracy DFT: Results from Applications of ‘*ab initio*’ QTP Functionals

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In an effort to emulate the high-accuracy results of coupled-cluster (CC) methods, while maintaining the computational efficiency and simplicity of density functional theory (DFT) methods, our group has developed a family of *ab initio* DFT functionals. These functionals are parameterized by enforcing an IP eigenvalue theorem on the one-particle exchange and correlation term of the Kohn-Sham energy operator. Previous works have shown that our QTP family of functionals can predict highly accurate values compared to CC and experiment for the IPs of small organic molecules (Ranasinghe, 2019) and the IPs/excited states of organic polymers (Windom, 2022). My recently published work demonstrates that, in addition to IPs, the QTP family can effectively predict EAs for a benchmark set of small organic molecules. QTP functionals consistently outperformed other standard range-separated and hybrid DFT functionals such as CAM-B3LYP and  $\omega$ B97x, with QTP02 and LC-QTP having MADs of  $<0.1$  eV. These findings support the idea that our IP-eigenvalue theorem for KS-DFT provides more accurate orbital eigenvalue solutions, and they are in line with the previous findings from accompanying studies. In addition, I have applied the QTP functionals to the first-order properties and excited state curves of the transition metal hydrides CuH and NiH, for which initial results show good agreement with CC and experiment. Further, layering the GW approximation onto QTP functionals for this problem reduces the IP and EA MADs to  $<0.2$  eV. My results, coupled with previous and ongoing work, demonstrate the wide variety of problems for which the QTP family of DFT functionals can be an efficient and accurate alternative to CC.