

Molecules as Neural Networks

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Given a quantum mechanical electron density for a molecule, an atom-in-molecule (AIM) decomposition partitions the molecular system into constituent atomic-like entities. This decomposition is not unique, and various theoretical methods have been proposed for defining chemically-reasonable AIMs. We describe the use of machine learning techniques, specifically radial basis function neural networks (RBF-NNs), to analyze molecular electron density distributions as AIM DFT ensemble decompositions [1]. The RBF-NN is designed to directly represent the AIM ensemble framework in terms of weighted superpositions of electron densities for individual ground, excited and charge states of the constituent atoms [2]. The neural network optimizes the electron density weights, while enforcing physical constraints, to define the contributions of individual atomic states to the overall molecular density. Our approach relies on universal function approximation theorems for neural networks [3].

We present results for LiF as an exemplar of ionic bonding and discuss insights into different bonding regimes derived from the neural network representation. Remarkably, we find that the RBF-NN can detect the transition from the ionically-bound molecule to isolated neutral atoms as a function of internuclear separation, R , independent of the level of theory. This is despite the fact that the levels of theory used to train the neural network fail to detect the transition via the computed dipole moment. The location of the transition is shown to depend on the level of theory. DFT models more equipped to represent charge transfer, such as wB97XD [4], locate the transition closer to the highly accurate quantum chemical results of Varandas [5]. This suggests that the quantum mechanical bonding information is intrinsic to the molecular electron density and transcends the exchange-correlation energy model used to compute it.

The RBF-NN framework provides a deeper understanding of contributions of individual atoms and atomic states to molecular properties and interactions, as a function of molecular structure. This includes bond formation and breaking – transitioning from isolated neutral atoms to chemical bonds – and the development of reactive charge transfer potentials for dynamical simulations [6].

Support from the NSF and from NSF REU grant #PHY-1659618 is gratefully acknowledged. We thank the UNM Center for Advanced Research Computing (CARC) for computational support.

References

- [1] S.R. Atlas. “Embedding quantum statistical excitations in a classical force field,” *J. Phys. Chem. A*, **125**, 3760 (2021); K. Muralidharan, S.M. Valone, and S. R. Atlas. “Environment dependent charge potential for water,” arXiv:0705.0857 [cond-mat.mtrl-sci] (2007).
- [2] G. Amo-Kwao and S. R. Atlas, “Radial basis function electron densities with asymptotic constraints,” ms., University of New Mexico (2024).
- [3] K. Hornik, “Multilayer feedforward networks are universal approximators,” *Neural Networks* **2**, 359 (1989).
- [4] J.-D. Chai and M. Head-Gordon, “Systematic optimization of long-range corrected hybrid density functionals,” *J. Chem. Phys.* **127**, 084106 (2008); J.-D. Chai and M. Head-Gordon, “Long-range corrected hybrid density functionals with damped atom-atom dispersion corrections,” *Phys. Chem. Chem. Phys.* **10** (2008) 6615-20 (2008).
- [5] AJC Varandas, *J. Chem. Phys.* **131**, 124128 (2009).
- [6] C.M. Baxter, S.M. Valone, and S.R. Atlas. “Ensemble embedding functions for atomistic dynamics,” ms. (2024).