A Neural-Network-Based Selective Configuration Interaction Approach to Molecular Electronic Structure

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A novel method is presented for efficient *ab initio* calculations of molecules' ground and excited electronic states. Combining Hartree-Fock with a neural-network-based configuration interaction (NNCI) algorithm enables selective configuration interaction calculations that mitigate the exponential growth of the Hilbert space. Using our recently developed Python library SOLAX [1], a neural network classifier iteratively selects basis elements relevant for the targeted states, optimizing the many-body basis. Applied to the N_2 molecule, NNCI reproduces full configuration interaction (FCI) results obtained on nearly 10^{10} Slater determinants by using only 4×10^5 [2]. We find that rather than being hindered by combinatorial growth, NNCI benefits from increasing the number of single-particle degrees of freedom, providing a scalable alternative to standard truncation schemes. Future directions are committed to extending this approach to a multi-tier embedding scheme, thereby trying to enhance the accuracy of electronic structure calculations for surface reactions in the context of heterogeneous catalysis.

[1] Louis Thirion, Philipp Hansmann, and Pavlo Bilous, "SOLAX: A Python solver for fermionic quantum systems with neural network support," *SciPost Phys. Codebases* (2025)

[2] Yorick L. A. Schmerwitz, Louis Thirion, Gianluca Levi, Elvar Ö. Jónsson, Pavlo Bilous, Hannes Jónsson, and Philipp Hansmann, "A Neural-Network-Based Selective Configuration Interaction Approach to Molecular Electronic Structure," *Journal of Chemical Theory and Computation* (2025)