A Compressed Sensing Approach to Select Optimal Atom-Centered Basis Functions for DFT and Beyond CHENCHEN WANG, Fritz Haber Institute, NIKLAS MENZEL, TU Berlin, LUCA M. GHIRINGHELLI, Fritz Haber Institute, GITTA KUTYNIOK, TU Berlin, MATTHIAS SCHEFFLER, Fritz Haber Institute — The choice of the basis sets is one of the most important factors in quantum chemical calculations. It is particularly challenging for functionals that treat electron correlations. Commonly used basis sets for advanced exchange-correlation functionals are not sufficiently accurate. This leads to extended basis set, such as the most famous correlation-consistent basis sets by Dunning. However, such basis sets have been so far widely used mainly for light atoms and their molecules, since they are too expensive for transition metals. We have developed a new approach to select basis functions based on compressed sensing (CS), a recently developed signal processing technique. CS provides a simple and efficient framework for basis selection based on l_1 norm regularization techniques. As introductory example, we select via our CS-based approach Gaussian basis functions (GTO) from a large pool of various GTOs, in order to fit to the reference atomic orbitals. We calculate the total energy for atoms from H to O, and then extend to molecules, e.g., H_2, N_2, and O_2. For H, He, and Li, our total-energy results are within 0.05% compared with STO-6G energies. Starting from Be, CS selected basis set provide significantly better results than STO-6G, even when only 5 GTOs are considered. Our new approach enables us to determine optimal basis sets for heavier atoms and molecules.

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