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Improving Density Functionals with Quantum Harmonic Oscillators

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Density functional theory (DFT) is the most widely used and successful approach for electronic structure calculations. However, one of the pressing challenges for DFT is developing efficient functionals that can accurately capture the omnipresent long-range electron correlations, which determine the structure and stability of many molecules and materials. Here we show that, under certain conditions, the problem of computing the long-range correlation energy of interacting electrons can be mapped to a system of coupled quantum harmonic oscillators (QHOs). The proposed model allows us to synergistically combine concepts from DFT, quantum chemistry, and the widely discussed random-phase approximation for the correlation energy. In the dipole limit, the interaction energy for a system of coupled QHOs can be calculated exactly, thereby leading to an efficient and accurate model for the many-body dispersion energy of complex molecules and materials. The studied examples include intermolecular binding energies, the conformational hierarchy of DNA structures, the geometry and stability of molecular crystals, and supramolecular host–guest complexes (A. Tkatchenko, R. A. DiStasio Jr., R. Car, M. Scheffler, Phys. Rev. Lett. 108, 236402 (2012); R. A. DiStasio Jr., A. von Lilienfeld, A. Tkatchenko, PNAS 109, 14791 (2012); A. Tkatchenko, D. Alfe, K. S. Kim, J. Chem. Theory and Comp. (2012), doi: 10.1021/ct300711r; A. Tkatchenko, A. Ambrosetti, R. A. DiStasio Jr., arXiv:1210.8343v1).