All-electron, Periodic GW and RPA Methods Within a Numeric Atom-centered Basis Set Framework

Xinguo Ren

CAS Key Laboratory of Quantum Information

University of Science and Technology of China, China

Email: renxg@ustc.edu.cn

Abstract:
We have implemented the periodic G^0W^0 method for quasiparticle energy calculations and the random-phase approximation (RPA) for ground-state total energy calculations within the all-electron, numerical atomic orbital (NAO) basis-set framework. A straightforward implementation of such correlated methods within the NAO framework is prohibitively expensive in terms of both the memory demand and CPU times. Here we employed a localized variant of the resolution of identity (RI) approximation, enabling a significant reduction of the computational cost to evaluate and store the two-electron Coulomb repulsion integrals. We demonstrate that the error arising from localized RI approximation is controllable and can be made negligibly small by enhancing the set of auxiliary basis functions (ABFs) used to expand the products of two single-particle NAOs. An efficient algorithm, suitable for the NAO framework, has been developed to deal with the Coulomb singularity in the Brillouin zone sampling. We performed systematic convergence tests and identified a set of computational parameters (basis sets, enhanced ABFs, and k-point grid), with which reliable G^0W^0 and RPA results can be obtained. Our implementation is carried out within the all-electron, NAO-based software package – the FHI-aims code [1,2]. Benchmark results for 3-dimensional insulators/semiconductors as well as 2-dimensional materials will be presented.

[1] V. Blum, F. Hanke, R. Gehrke, P. Havu, V. Havu, X. Ren, K. Reuter, and M. Scheffler, Comp. Phys. Comm. 180, 2175 (2009).
[2] X. Ren, P. Rinke, V. Blum, J. Wieferink, A. Tkatchenko, A. Sanfilippo, K. Reuter, and M. Scheffler, New J. Phys. 14, 053020 (2012).