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Relativistic GW for solids and molecules

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The fully self-consistent GW (scGW) method with an iterative solution of the Dyson equation provides a consistent approach for describing the ground and excited states without any dependence on the mean-field reference. In this work, I will discuss a relativistic version of scGW for molecules and solids containing heavy elements using the exact two-component (X2C) Coulomb approximation. We benchmarked the SOC-81 data set containing closed shell heavy elements for the first ionization potential using the fully self-consistent GW as well as one-shot GW. The self-consistent GW provides superior results compared to G0W0 with PBE reference while removing the starting point dependence. The photoelectron spectra obtained at the X2C level demonstrate very good agreement with the experimental spectra. We also observe that scGW provides very good estimation of ionization potential for the inner d-shell orbitals. Additionally, using the total energy, we investigate the equilibrium bond length and harmonic frequencies of a few halogen dimers using scGW. Overall, our findings demonstrate the applicability of the fully self-consistent GW method for accurate ionization potential, photoelectron spectra, and total energies in finite systems with heavy elements with a reasonable computational scaling. Finally, I will discuss X2C-1e scGW calculations for solids such as semiconductors/metals, conventional insulators, and topological insulators. Here we will analyze conventional properties such as band structure, equilibrium lattice constant and bulk modulus in addition to discussing the zeros of Green's functions and leading a novel analysis applicable to discovering properties of novel topological materials.

References

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