Route to control precision in accurate many-body methods in the predictions of electronic and optical properties of 2D materials.

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Abstract:
Achieving precise and accurate predictions of the electronic and optical properties of materials is a critical task in materials science. To this end, computationally expensive many-body methods such as GW perturbation theory and the Bethe-Salpeter equation have been developed. However, different materials have varying levels of sensitivity to the technical parameters required to achieve precise values for electronic band gap, exciton binding energy, and absorption spectra. Carefully converged results from these methods are then directly comparable with experiments[1] or with fixed-node diffusion Monte Carlo[2] (Figure 1). In this study[3], we investigate the sensitivity of these parameters on a set of III-V binary hexagonal semiconductors and semiconducting MXenes and propose an explanation as to why certain parameters are more important in some materials than others. Our analysis includes real-space projections of excitonic wave functions (Figure 2) and partial electron densities, with the size and orientation of these entities corresponding to the observed trends. Our results provide insight into the complex interplay between material properties and computational methods and have important implications for the design and optimization of novel 2D materials.

References

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[2] Dubecký M., Minárik S., Karlický F.: Benchmarking fundamental gap of $Sc_2C(OH)_2$ MXene by many-body methods. *J. Chem. Phys., 158, 054703 (2023)*
[3] Kolos M., Karlický F.: Electronic and Optical Properties of III-V Binary 2D Semiconductors: How to Achieve High Precision from Accurate Many-Body Methods. *Phys. Chem. Chem. Phys., 24(44), 27459-27466, (*2022)

Figures

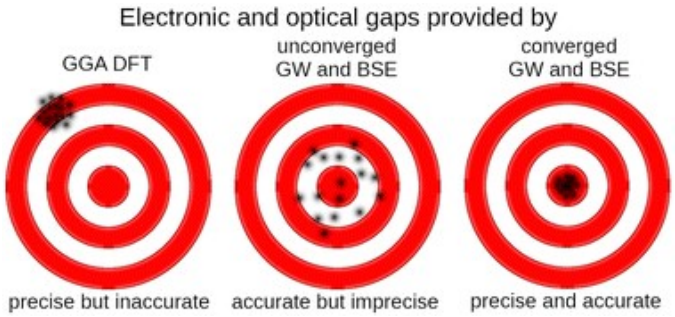

Figure 1: Precision and accuracy in density functional theory and many-body methods.

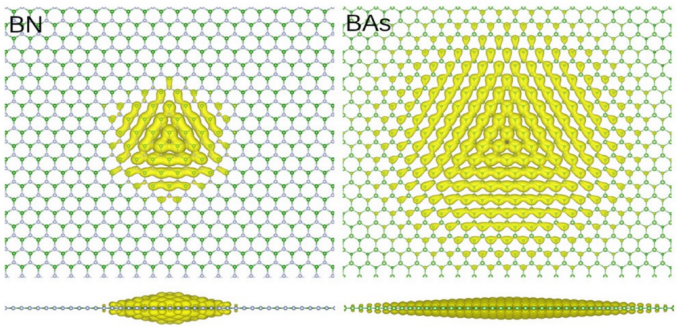

Figure 2: Top and side views of the isosurface of the first bound exciton with a fixed hole for BN (left) and BAs (right) plotted with the same isosurface level. The size of the first exciton influences optical gap convergence.