

Catchy Title (brief)

Electronic and Optical Properties of Bilayer α - B_2N_2 in Different Stacking Modes

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In Short

- Previous PBE, G_0W_0 @PBE and BSE results show strong stacking dependence of quasiparticle band gaps, excitonic behavior, and visible-light optical response.
- For some stackings (AA, AA' and AB''), the PBE starting point exhibits semimetallic or near-zero-gap character, leading to unphysical screening and band distortion near the Y point in G_0W_0 @PBE calculations.
- Adopting HSE as the new mean-field starting point and performing G_0W_0 @HSE and BSE calculations is therefore essential for obtaining trustworthy quasiparticle and optical properties.

Bilayer orthorhombic boron nitride (α - B_2N_2) is a promising two-dimensional optoelectronic material whose electronic structure and optical response are highly sensitive to the interlayer stacking arrangement. As shown in Figure 1, this project systematically investigates six representative bilayer stacking configurations. In the previous stage, we examined the quasiparticle band structures, excitonic effects, and optical properties of bilayer α - B_2N_2 for all six stackings based on PBE, G_0W_0 @PBE, and the Bethe-Salpeter equation (BSE). The results show that all stacking configurations retain a direct-gap character at the quasiparticle level, with band gaps ranging 0.67-1.99 eV, compared with the results obtained from semilocal density-functional calculations, the G_0W_0 band gaps are significantly enlarged, with an increase of about 1.3 eV, and their quasiparticle band gaps, exciton types, and optical responses in the visible-light region exhibit pronounced stacking dependence. These results establish an initial physical picture of the stacking-dependent electronic and optical properties of bilayer α - B_2N_2 .

However, the previous study also revealed a decisive theoretical bottleneck. For some stacking configurations, the PBE starting point yields a semimetallic or near-zero-gap character, which introduces unphysical metallic screening in the subsequent G_0W_0 calculations and leads to noticeable band distortions near the Y point in the Brillouin zone, as shown in Figure 2. Since the band-edge electronic structure directly determines the quasiparticle band gap, excitonic transition channels, and the position of

the optical absorption onset, such an inaccuracy in the starting point further affects the quantitative interpretation of excitonic properties and optical responses.[1] Therefore, PBE is not a sufficiently reliable many-body starting point for this system, and relying solely on G_0W_0 @PBE is insufficient for obtaining publication-quality and quantitatively trustworthy results.[2]

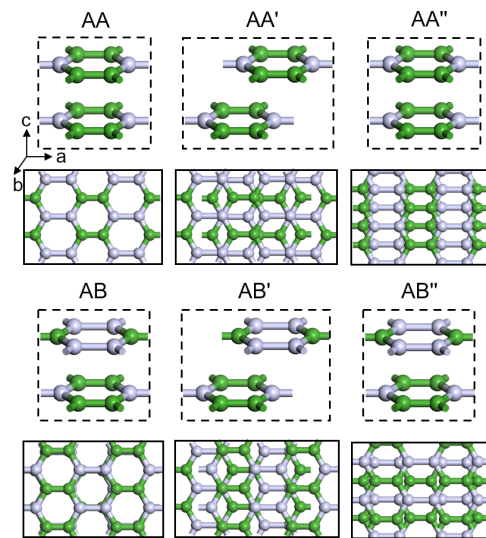


Figure 1: Side (top) and top (bottom) view of six stacking structures of bilayer α - B_2N_2 .

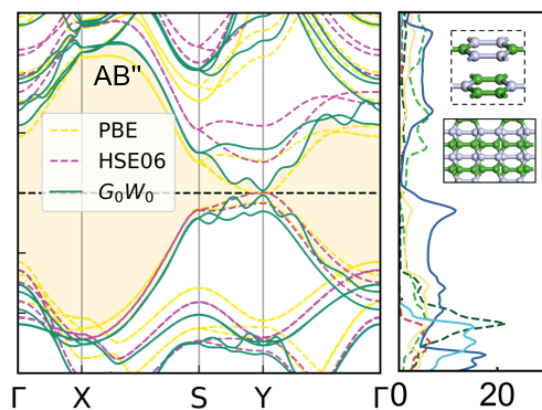


Figure 2: Example of the quasiparticle band-structure distortion obtained from G_0W_0 @PBE for a representative AB' stacking configuration. The distortion appears near the Y point in the Brillouin zone, reflecting the unphysical screening effect introduced by the semimetallic or near-zero-gap PBE starting point.

Based on the above understanding, adopting the hybrid functional HSE as a new mean-field starting point is a necessary key methodological step for obtaining physically reliable many-body results

in bilayer o-B₂N₂. The calculations in the previous stage have already shown that, for some stacking configurations (AA, AA' and AB''), the PBE starting point yields a semimetallic or near-zero-gap character, and introduces anomalous metallic screening in the subsequent G₀W₀ calculations, ultimately leading to nonphysical distortions in the quasiparticle band structure. Therefore, the uncertainty in the current results does not simply originate from ordinary numerical errors, but rather from the intrinsic limitation of PBE itself as the starting point for the many-body calculations.[3]

In this context, the introduction of HSE is clearly necessary and irreplaceable, because it can provide a more realistic description of the single-particle electronic structure, especially the band-edge-state distribution and the finite-gap character of the relevant stacking configurations.[4] By correcting the unreasonable semimetallic tendency given by PBE already at the mean-field level, HSE is expected to suppress the anomalous screening effect at its origin and thereby remove the nonphysical band distortion observed in G₀W₀@PBE. This is crucial, because the band-edge states directly determine the quasiparticle band gap, excitonic transition channels, and the position of the optical absorption onset. Without a reliable starting point, even if the subsequent G₀W₀ and BSE calculations can capture the general trends, the results can hardly be regarded as quantitatively trustworthy.

Therefore, the present project extension is a necessary continuation aimed at overcoming the core theoretical bottleneck already clearly identified in the previous stage. The central goal of the extension phase is to establish a trustworthy quasiparticle and excitonic physical picture for bilayer o-B₂N₂ by carrying out G₀W₀@HSE and BSE calculations. These calculations are indispensable for eliminating the current band-distortion problem, obtaining quantitatively reliable electronic-structure and optical-spectrum results, and ultimately producing high-quality results suitable for submission to *Physical Review B*.

[3] W. Chen, A. Pasquarello, *Phys. Rev. B* **90**, 165133 (2014). doi:10.1103/PhysRevB.90.165133

[4] Stephen E. Gant, Jonah B. Haber, Marina R. Filip, Jeffrey B. Neaton, *Phys. Rev. Mater.* **6**, 053802 (2022). doi:10.1103/PhysRevMaterials.6.053802

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More Information

[1] R. Rodrigues Pela, C. Vona, S. Lubeck, B. Alex, I. Gonzalez Oliva, C. Draxl, *npj Comput. Mater.* **10**, 77 (2024). doi:10.1038/s41524-024-01253-2

[2] R. Rodrigues Pela, U. Werner, D. Nabok, C. Draxl, *Phys. Rev. B* **94**, 235141 (2016). doi:10.1103/PhysRevB.94.235141