Ab Initio Modelling of Two-Dimensional Semiconductors

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Introduction

- Well-known, popular modelling approaches like Density Functional Theory (DFT) are often **inadequate** for the study of condensed matter systems.
- Quantum Monte Carlo (QMC) techniques are a family of alternative methods for the study of a wide variety of condensed matter systems [1], offering **systematic improvement** over other techniques.

Variational MC

- Simplest of all QMC techniques, reliant on variational principle for the **trial many-e$^-$ wavefunction** $|\Psi_\alpha\rangle$:

$$E(\alpha) = \frac{\langle\Psi_\alpha|\hat{\mathcal{H}}|\Psi_\alpha\rangle}{\langle\Psi_\alpha|\Psi_\alpha\rangle} \geq E_0 \qquad (1)$$

  $\alpha \longrightarrow$ variational parameters, chosen **specially**
  $\hat{\mathcal{H}} \longrightarrow$ many-e$^-$ Hamiltonian
- VMC used as prelude to Diffusion MC (DMC) studies. Cheaper to add **variational freedom** and exploit it *before* using more computationally expensive DMC.

DMC - Biexcitons in Coupled QWs

- Two 2DEGs, separated by d. Already realised in III-V bilayers, *similar*† physics seen in **truly** 2D heterostructures.

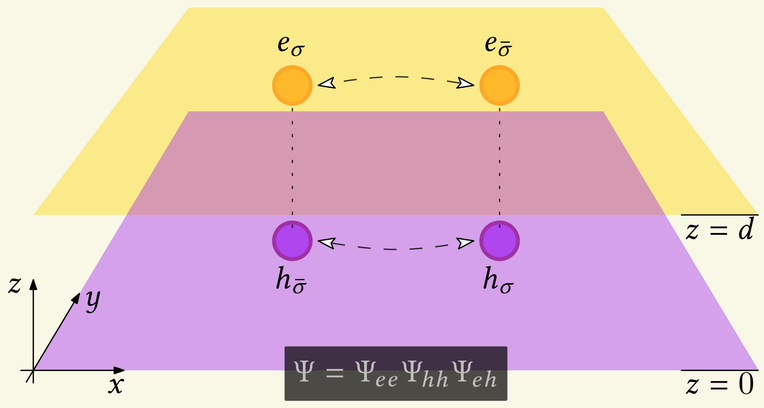


Figure 1: Schematic of biexciton formation in a CQW system. Inset: form of the biexciton trial wavefunction.

- DMC is a **projection** method, $|\Psi\rangle$ evolved in *imaginary* time ($\tau = it$)

$$|\Psi(\tau)\rangle = \sum_{i=0}^{\infty} c_i \, \mathrm{e}^{-(\epsilon_i - E_T)\tau} |\Phi_i(0)\rangle \qquad (2)$$

- e^-h^+ & $e^-h^+e^-h^+ \longrightarrow$ **nodeless** model systems

† **Problem**: screening (like [2]) in realistic systems...

E_{DFT}

E_{VMC}

E_{DMC}

E_0

DMC - hBN energy gaps

- QMC methods aren't just for toy problems

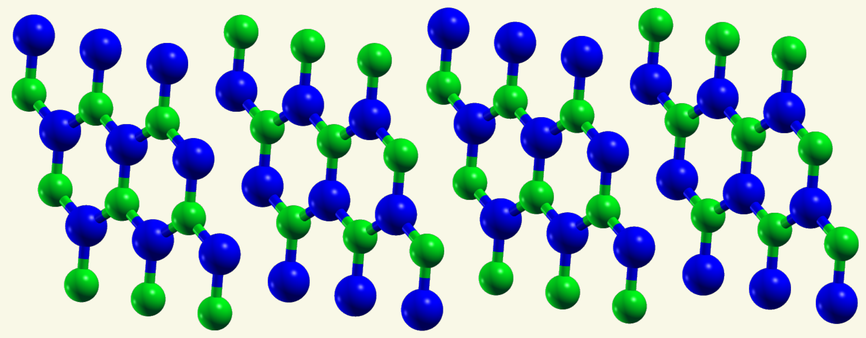
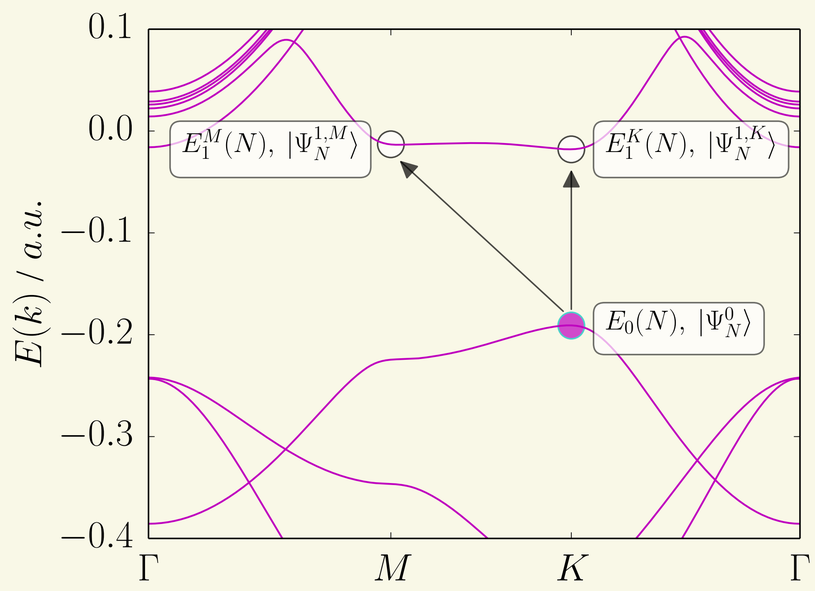


Figure 2: An example (3 3 2) supercell used in our simulations. Blue atoms are B, Green are N. This sim. cell contains 144 e$^-$ (PPs!).

- **Goal:** Use QMC to calculate, from first-principles, the *excitonic* and *quasiparticle* gaps of bulk hBN,

$$\Delta^P_{Ex} = E^P_1(N) - E^P_0(N) \qquad (3)$$

Figure 3: Example *monolayer* hBN DFT (PBE) bandstructure, labelled for explanation.



- **Why?** $P \in \{K, M, \cdots\} \implies$ can *accurately* determine gaps between arbitrary points in $\vec{k}$-space..
- **Progress?** Ask me for a real-time update!

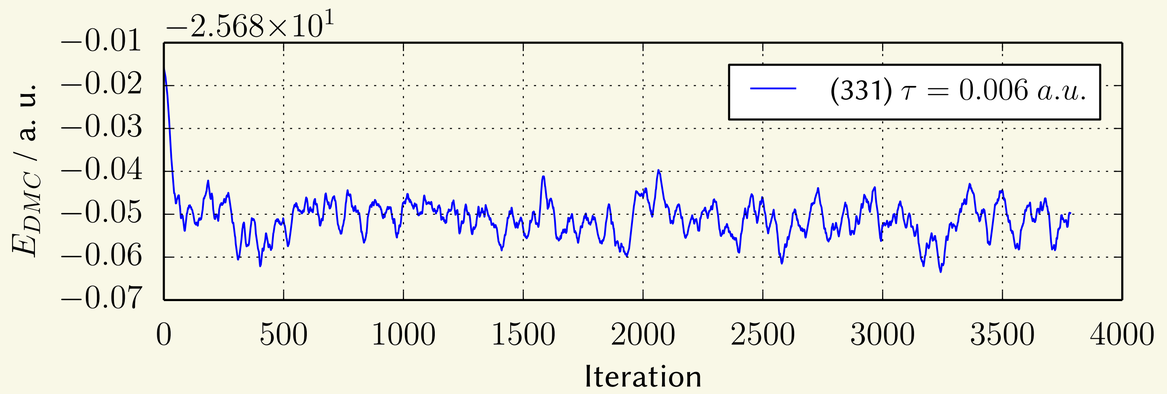


Figure 4: My first results, for g.s.e of the (3 3 1) supercell.

Acknowledgements

- QMC calculations performed with the CASINO code [4].
- All calculations were performed on the **HEC** facility at Lancaster and the **N8 HPC**.

References

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