Modelling high-precision TEM phase imaging with DFT

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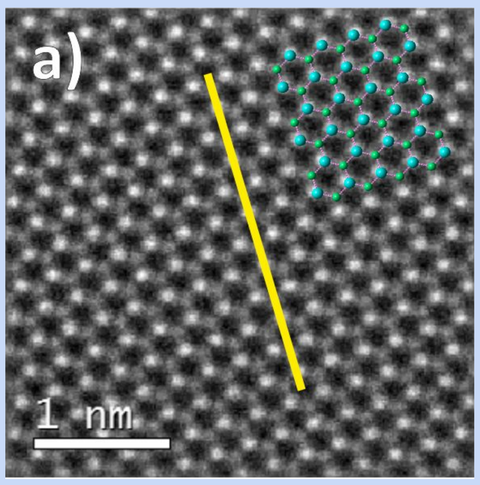
OxfordMaterials

ELECTRON MICRSCOPY AND SIMULATION

- Scanning transmission electron microscopy (**STEM**) create images at an **individual atom resolution.** Quantum mechanical simulations are necessary to be able to fully interpret these experimental results.
- The conventional independent atom model (**IAM**) uses precalculated projected potentials of isolated atoms superimposed into the required crystal lattice. Subsequent image includes **no charge transfer effects** between atoms.
- **Ptychography [1] (image generation from coherent interference patterns)** applied to STEM provides images which are sensitive to both light and heavy elements - a challenge for other techniques.
- Due to this added precision, there is a measurable difference between IAM simulation images, and experiment - **due to lack of charge transfer**.
- It is possible to utilise CASTEP [2], a DFT code to calculate a projected potential for a crystal which **includes charge transfer**.
- A monolayer of hexagonal boron nitride (h-BN) is being used for this project, due to there being significant charge transfer.



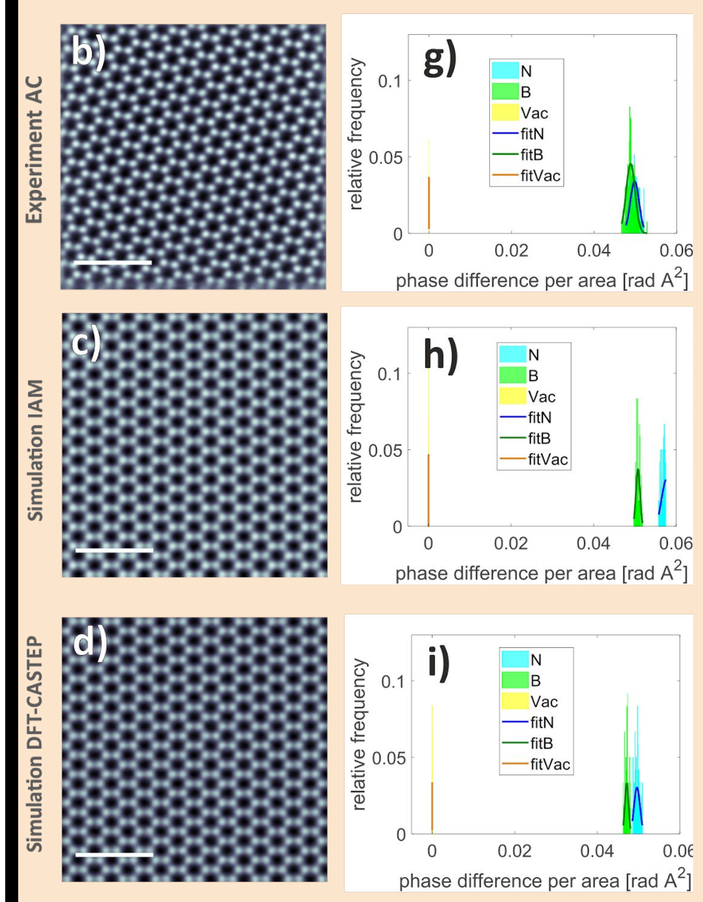
Experimental image of monolayer of hBN, taken from [3]

- Previous results have modified CASTEP to produce projected potentials, which was used to generate phase images which improved against IAM when compared to experiment. However, gap exists between DFT and experiment; efforts will be made to close the gap.

PROJECT

- Due to the early nature of this project, this poster will focus on demonstrating the difference in results between IAM and the DFT code.
- Then, a summary of the work in progress, plus future plans will be shown.

PHASE IMAGES

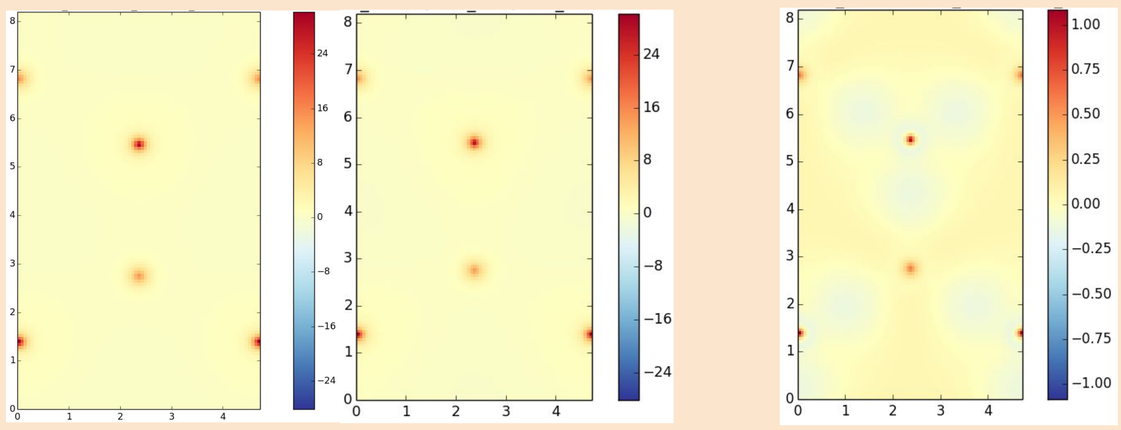


Selected relevant figures from [3]. Ptychographic phase image (b), simulated IAM phase image (c), simulated DFT phase image (d). g), h), i) contain respective phase differences per area.

- In order to better distinguish between the phase image, a histogram for each plot showing the phase difference per area vs relative frequency has been included.
- The phase difference histogram for experiment demonstrates N and B have overlapping peaks, which is not found in IAM simulation.
- This is mostly corrected in DFT simulation, but two distinct peaks are still visible.

- In conclusion, there is a smaller, but still discernible gap between simulation and experiment for h-BN.
- Now ptychography applied to STEM has allowed for high-intensity phase images which capture charge transfer effects, a more sophisticated simulation approach needs to be developed.

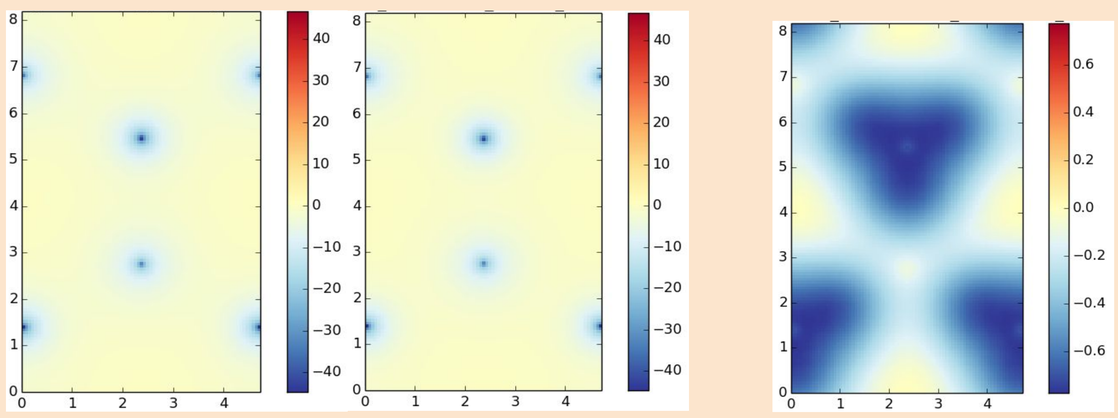
ELECTRON CHARGE DENSITY COMPARISON



h-BN charge density from IAM (left) and h-BN charge density from DFT (middle), with IAM - DFT(right).

- IAM overestimates the charge density at the nucleus, and underestimates the charge density around the nucleus.
- **Electrons are shown to relax, which demonstrates charge transfer from B to N**.

PROJECTED POTENTIAL COMPARISON



h-BN projected potential from IAM (left) and h-BN projected potential from DFT (middle), with IAM - DFT(right).

- Looking at the projected potentials emphasises the differences found by looking at the charge density.
- **The projected potential surrounding N is screened**.

WORK IN PROGRESS

- Further development is required in order to improve simulation. The code currently neglects effects due to **phonons.** A possible approach is to implement via frozen phonons:
  - Calculate the temperature dependent available phonon modes.
  - As with experiment, each probe position will measure the nucleus being at a different position, which will depend on which mode the nucleus is in, and which position it happens to be in during measurement.
  - The subsequent charge density, projected potential, and phase image will therefore be different and should better reflect experiment
- So that the code is useful for a variety of systems, it is being made parallel.

FUTURE PLANS

- All results were collected using the **PBE functional**. It may or may not be the most appropriate - there will be investigations to see if this is the source of the existing gap.
- Currently the code only works with 2D materials. By propagating the electron beam iteratively through each slice, **the multislice method**, the code will be able to deal with 3D, bulk materials.
- Generate the phase images with an in-house code.

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References

[1] H. Yang, R. N. Rutte, L. Jones, M. Simson, R. Sagawa, H. Ryll, M. Huth, T. J. Pennycook, M. L. H. Green, H. Soltau, Y. Kondo, B. G. Davis, and P. D. Nellist. Simultaneous atomic-resolution electron ptychography and z-contrast imaging of light and heavy elements in complex nanostructures. Nature Communications, 7:12532 EP –, 08 2016.

[2] M D Segall, Philip J D Lindan, M J Probert, C J Pickard, P J Hasnip, S J Clark, and M C Payne. First-principles simulation: ideas, illustrations and the castep code. Journal of Physics: Condensed Matter, 14(11):2717, 2002.

[3] G T Martinez, T C Naginey, L Jones, C O'Leary, T J Pennycook, R J Nicholls, J R Yates, and P D Nellist. Direct imaging of charge redistribution in monolayer hexagonal boron nitride using electron ptychography in the scanning transmission electron microscope. In preparation.