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Computational Materials Modelling

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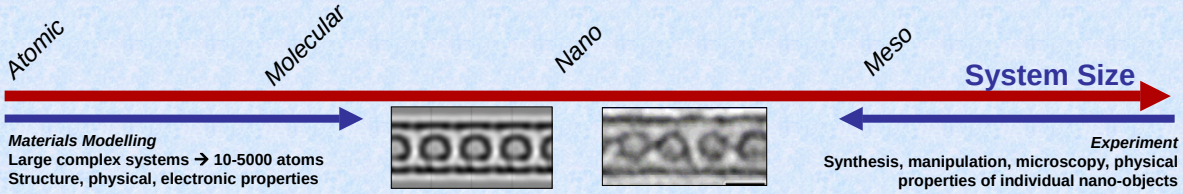
Motivation

Modern materials modelling techniques allow both interpretation **of experimental data and** prediction **of new materials properties. Improvements in nanoscale synthesis and characterisation are bringing experimental techniques towards quantitative analysis at the nanoscale. At the same time, improvements in computational modelling methods allow us, for the first time, to directly correlate atomic structural models with experiment.**



Approach

First Principles Calculations: Using computer software to solve the fundamental equations of Quantum Mechanics, we can predict materials behaviour with no input from experiment. These techniques are the most accurate but are limited in the complexity of system that can be simulated.

→ ***Techniques***: Density Functional Theory (DFT) via the AIMPRO (Newcastle), TB-LMTO (Stuttgarde) and SIESTA (Spain) codes.

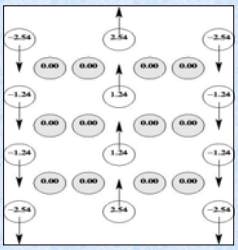
Semi-empirical Calculations: These use parameters fitted to experiment and/or to first principles data in order to reduce the computational cost. This allows to study more complex and extended systems.

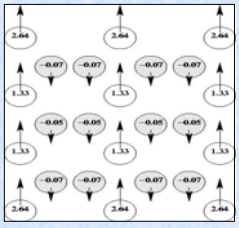
→ ***Techniques***: TB-RS, DFTB (Dresden)

These two approaches are complementary to interpret experimental observations and predict new properties of materials.

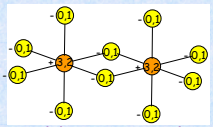
Examples

Semiconductor / ferromagnetic interfaces.
Magnetic moments of the 4 Fe monolayer and 3 Ge monolayer film in the antiferromagnetic (left) and ferromagnetic (right) configurations. Ge atoms are shown as filled circles and Fe by empty circles. The values of the local magnetic moments indicated inside the circles are in μ_B.

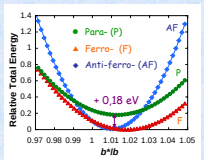


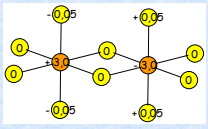


Ferromagnetic (F) and antiferromagnetic (AF) configurations obtained by TB-LMTO calculations in jamesonite ($FePb_4Sb_6S_{14}$) where the magnetic atoms (Fe, in red) are surrounded by sulphur atoms (yellow) forming a 1D-like structure.



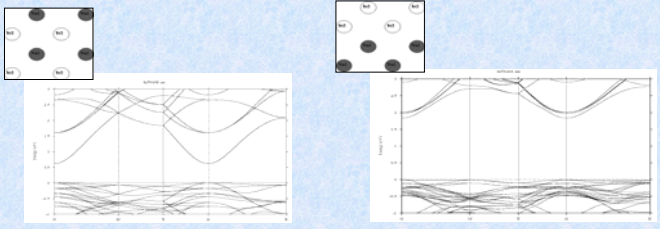
(F) Ferromagnetic



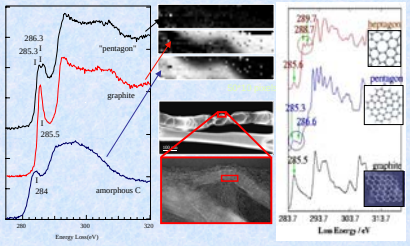


(AF) Anti-ferromagnetic

Band structure calculations to understand the evolution of the optical band gap of photovoltaic materials.
This figures show the very sensitive effect of the Na atoms rearrangement in the $NaIn_5S_8$ ordered compound.



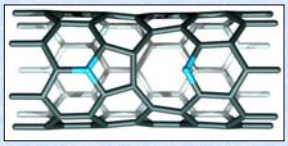
EELS of defective nanotubes



Experimental EELS — Theoretical EELS

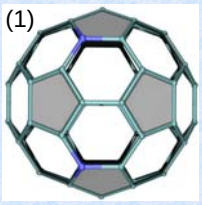
(Simulations in collaboration with Sussex and Paris Sud)

Nitrogen doped carbon nanotubes



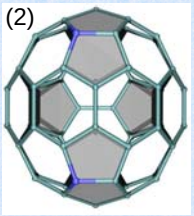
(7,0) carbon nanotube, two nitrogen atoms along the axis (blue), with single vacancy between them. Vacancy formation energy <2.5eV (normally >5eV).

Azafullerenes : Nitrogen violation of the isolated pentagon rule

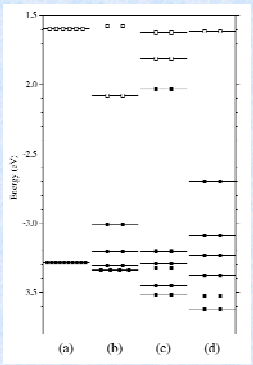


$C_{58}N_2$

(2) contains two pentagon pairs, and is more stable than (1) by 0.54eV. In C_{60} (2) is 1.6eV less stable.



Nitrogen stabilises the paired pentagons, and violates the isolated pentagon rule



DFT Kohn-Sham eigenvalues (eV) at the HOMO-LUMO gap

(a)C_{60}
(b)C_{60} with one bond rotation (2 × paired pentagons)
(c)$C_{58}N_2$, structure as (a)
(d)$C_{58}N_2$ as (b) with one N atom per paired pentagon.

We predict a **new family** of nitrogen doped fullerene molecules.

Collaborations

- **IPCMS Strasbourg University ,** C. Demangeat
- **Valladolid University,** A. Vega

References

Optical Materials 2005, 27, 647-653

Surface Science 2005, 576, 158-164

Collaborations

- **European STREP (nano2hybrids) 2007** Namur, Louvain-La-Neuve, Vega Science Trust (Sussex), ULB Brussels, Sensotran (Spain)
- **Sussex University (UK)** M. Heggie, H. Kroto
- **Université Paris Sud** C. Colliex, A. Gloter, O. Stephan
- **IJS, Slovenia** D. Arcon, P. Umek
- **University of Oslo (Norway)** M. Glerup

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'Nitrogen Doping in Carbon Nanotubes' **Invited Review, C. Ewels**, M. Glerup, **J. Nanosci. Nanotech.**, 5 (9), 1345-1363 (2005).

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Future work: Increasingly complex systems, such as PPV-polymer-nanotube interactions, bio-nano composites, complex multi-function hybrid nanomaterials (oxides, magnetic minerals)

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