Modelling Nano Peapods for Quantum Computing
Dr Ling Ge, Department of Materials, Oxford University

Nanotechnology is the engineering of materials, devices and operating systems on a very small scale, typically within the range of just under 1 and up to 100 nanometres (nm). One nanometre is equal to one billionth of a metre (1×10^{-9} m). To put that in context, the smallest existing pollen grain is 6 micrometres in diameter, 6,000 times the size of a single nanometre. While a single carbon atom has a diameter of about 0.1nm, buckminsterfullerenes, which are hollow, spherical structures composed of between 20 and 60 carbon atoms, have a diameter of about 0.7nm. We can also put atoms inside the carbon buckyball, (the so-called endohedral fullerene). Once we are looking at systems containing a few hundred atoms ~10nm across we can't avoid quantum effects!

The use of nanotechnology has witnessed explosive growth in recent years, finding use in medicine (nanomedicine), electronics, consumer products and nanomaterials. The application of nanoparticles in sunscreens is one of the most common uses in consumer products today: well over 300 sunscreens on the market contain zinc oxide and titanium dioxide nanoparticles to reduce the visibility of the cream. Silver nanoparticles can be added to fabrics to kill bacteria and make them odour resistant. Nanotechnology is able to reduce the size and lower the production and operation costs of devices such as solar cells, fuel cells and batteries.

A much more recent application of nanotechnology is in the development of quantum computing: an entirely new form of computer technology, which leaves behind classical mechanics and instead uses principles based on quantum theory. Quantum theory is essentially based on the idea that as objects become extremely small (on the atomic and molecular scale), they start to behave very differently to how we usually know and understand. You may have heard of the concept of wave-particle duality, which explains that very small particles, such as electrons can behave as both a particle, with mass and momentum, and a wave. It may be helpful to imagine a travelling electron as a particle, whose path is controlled by a wave. The same idea can be applied in quantum computing.

As you may have heard from newspapers and televisions, quantum computing has become a fascinating research area nowadays. In particular, I am going to talk about single walled carbon nanotubes (SWNT), which have things inside them that might be used to embody the quantum information. Spin chains have potential to provide the controlled interactions needed for quantum computing. Carbon is a candidate host for spin quantum bits (qubits) because in ^{12}C materials the small spin-orbit coupling and absence of hyperfine coupling ensures long spin coherence times. Carbon peapods, that is, single-walled carbon nanotubes containing fullerenes, have been proposed as particularly suitable spin chain systems. The fabrication of nanoscale electronic devices, such as field effect transistors, with carbon peapods containing various endohedral fullerenes is well established. When metallic atoms with an unpaired electron such as scandium (Sc) are incarcerated in a carbon cage, the system develops hybridised orbitals resulting in an unpaired electron delocalised across the fullerene cage – potentially a near ideal qubit.

Here we report on detailed numerical simulations that establish the nature of the spin-spin interactions both between endohedral fullerenes and between fullerenes and nanotubes. The dominant interaction is of the Heisenberg form, which has potential for quantum computing in the one-dimensional chain. Quantum computing protocols have been devised and experimentally demonstrated which allow chains of identical units to be controlled globally,

circumventing the difficulty in local addressing. For this to be achieved it is necessary to set and measure the qubits at the end of the chain, but all the other qubits in the chain can be manipulated collectively, without the need for individual addressing. In this way information can be propagated and processed in the spin chain, and scalable quantum computations can be performed.

Four fundamental problems need to be understood in order to demonstrate well-defined qubits in carbon peapods: (i) the charge arrangement within the carbon peapods; (ii) the electron spin distribution; (iii) the coupling between spin-qubits; (iv) the nature of the spin interactions between fullerenes and nanotube.

The CRYSTAL code, developed by STFC's Computational Materials Science Group with collaborators at the University of Turin, was used to perform these *first principles* calculations. We find well-defined spin-1/2 qubits on the fullerenes, with strong evidence for a nearest-neighbour Heisenberg exchange interaction. In order to describe the influence on the spin-qubits localised on the fullerenes of propagating electrons or holes in the nanotube, it is necessary to go beyond density functional theory to a model which is capable of describing the low-energy charge-spin excitations of the system. We conjecture a generic Hubbard-Anderson model; which captures the Heisenberg exchange between spins along the fullerene chain and the Kondo exchange interaction between localised spins on the fullerenes and spins of propagating electrons or holes in the nanotube.

Figure 1 (a) shows the calculated electronic eigenspectrum for the relaxed $Sc@C_{82}$. Sc has three valence electrons and the ground state of $Sc@C_{82}$ is found to be a spin-1/2 system. The unpaired electron occupies the highest occupied molecular orbital (HOMO) of $Sc@C_{82}$, which constitutes the spin qubit. The HOMO is well separated from the energy levels above and below leading to a well-defined qubit. We establish that the HOMO of $Sc@C_{82}$ is virtually identical to the (LUMO+1) of C_{82}, as shown in Fig.1 (c), whereas the lower lying orbitals are hybrids of Sc and C_{82}. Thus Sc acts as a perfect donor to the C_{82} cage for the HOMO state.
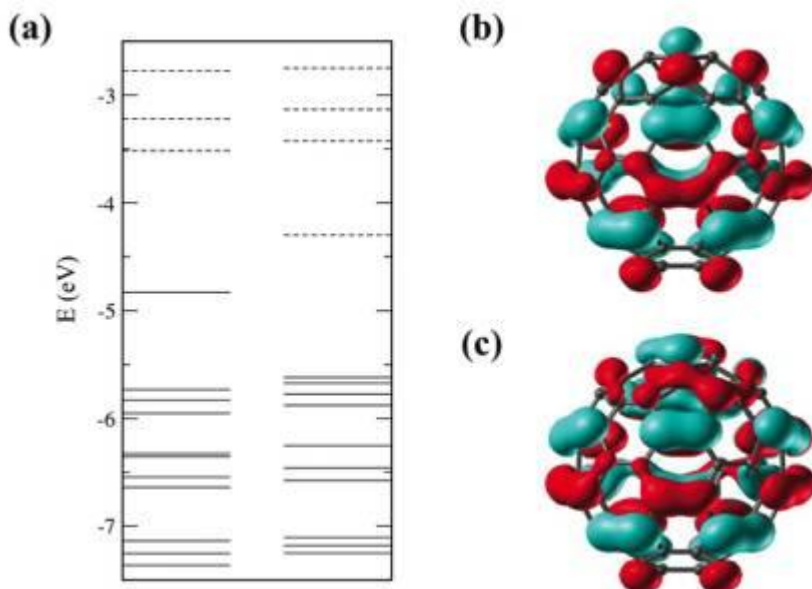

Figure 1 – Electronic structure of $Sc@C_{82}$

Figure 2 shows the electronic charge rearrangement following the $Sc@C_{82}$ encapsulation in the (14,7) nanotube (semiconducting). The charge depletion from the nanotube is concentrated around the fullerene sites. Similar qualitative results are obtained for the (11,11) nanotube (metallic). In both peapods, electron transfer occurs from the nanotube and

the Sc atom to the C_{82} cage due to hybridisation between the occupied states of the nanotube and fullerenes. The charge transfer from the Sc atom to the C_{82} cage in Sc@C_{82} @(14,7) and Sc@C_{82} @(11,11) is very similar to that of Sc@C_{82}. The shape of the spin density distribution in the peapods closely resembles that of the HOMO of Sc@C_{82} illustrated in Figure 1 (b). The charge transfer and spin distribution are insensitive to encapsulation.
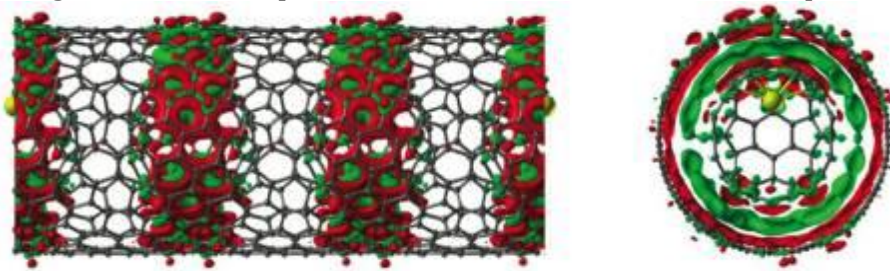

Figure 2 – Charge transfer in Sc@C_{82}@(14,7) peapod

In the predicted ground state configuration of the peapods, the spin direction alternates along the Sc@C_{82} chain; the corresponding configuration with parallel spins being higher in energy. We denote these configurations as antiferromagnetic (AF) and ferromagnetic (FM) states, respectively. Both states are found to be Mott insulators and have a total energy lower than the restricted Kohn-Sham solution. The exchange parameter *J*, defined as the energy difference between FM and AF configurations, is 3 meV per cell (containing two spins). The behavior of *J* as a function of the inter-fullerene separation, *R*, in a Sc@C_{82} chain is plotted in Figure 3. Values of *J* calculated for the peapods at discrete values of *R*, as indicated in Figure 3, coincide with those obtained for the Sc@C_{82} chain within the accuracy of the present calculations. At these separations the inter-molecular spin interaction is therefore via direct exchange between fullerenes, with a negligible contribution from interactions via the nanotube. This inter-molecular coupling is much larger than the classical dipole coupling of N@C_{60} and even larger than that computed for defective fullerenes with inter-cage links. This surprising result follows from the HOMOs in the Sc@C_{82} chain being very extended as illustrated in Figure 3. The p_z orbitals on the closest C atoms belonging to adjacent molecules overlap in a σ-type fashion. This implies that the exchange interaction could be tuned by varying the separation between the fullerenes in peapods.
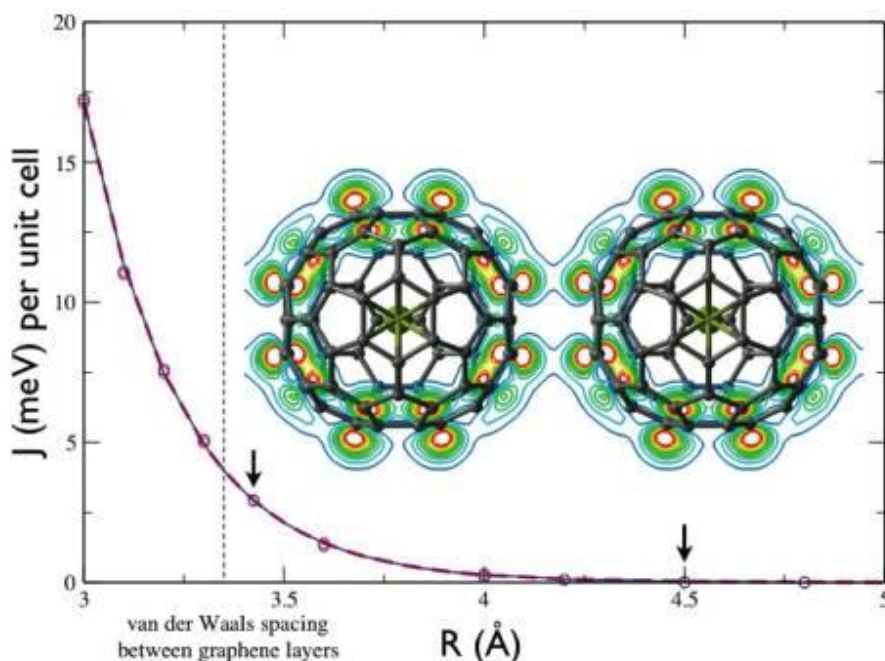

Figure 3 – Exchange interaction strength as a function of the interfullerene separation for an isolated chain of Sc@C_{82} fullerenes (full curve) and the peapod structures (arrows).

The AF band structures of the peapods are plotted in Figure 4. They are consistent with a chain of fullerenes interacting weakly with a nanotube. In the semiconducting case, Figure 4a, there are very narrow bands, consistent with weak hopping of electrons along the fullerene chain. In the metallic case, Figure 4b, these bands cross the wide nanotube bands. The weak interaction opens small hybridisation gaps (circled in Figure 4) that are reproduced in the generic Anderson-Hubbard type model.
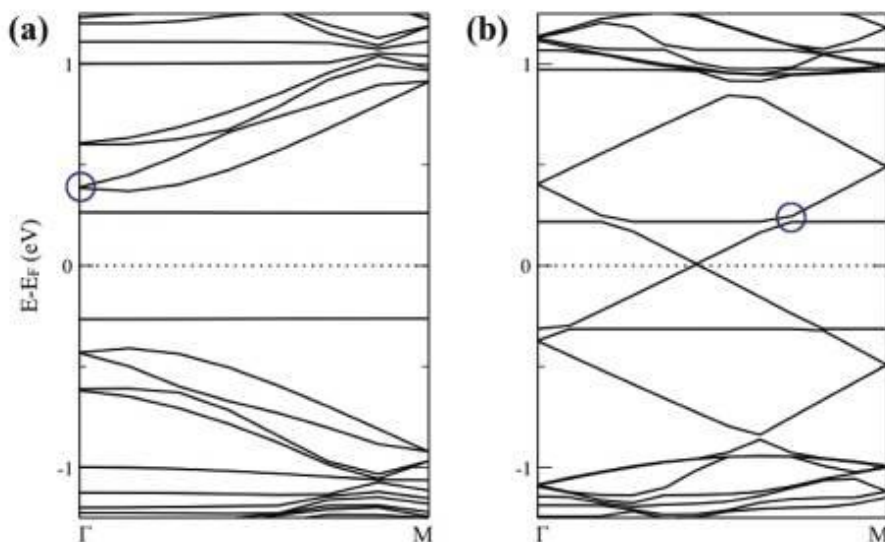

Figure 4 – Spin-polarised band structures of the (a) $Sc@C_{82}@(14,7)$ and (b) $Sc@C_{82}@(11,11)$ peapods for the AF configuration.

In conclusion, $Sc@C_{82}$ peapods offer well-defined spin-qubits on the C_{82} cage, coupled via antiferromagnetic Heisenberg exchange interactions. For the semiconducting case, the upper and lower Hubbard bands of the fullerene chain are little affected by the nanotube and occur entirely within the band gap of the nanotube, allowing excitations of the $Sc@C_{82}$ chain independently of the nanotube. Remarkably, for the peapods discussed in this paper, the main function of the nanotubes will be to give mechanical support for the endohedral fullerenes and to protect them from the environment, rather than to provide controlled interactions between the spins. An endohedral fullerene peapod thus provides a candidate nanostructure for spin-chain quantum computing.

★

Dr Ge's DPhil work has been published in several prestigious journals such as Nature Nanotechnology, American Chemical Society (ACS) Nano, Nano Letters, and Physical Review B.

References:
1. L. Ge, B. Montanari, J. H. Jefferson, N. M. Harrison, D. G. Pettifor, and G. A. D. Briggs, Phys. Rev. B **77**, 235416 (2008)
2. L. Ge, John H. Jefferson, B. Montanari, N. M. Harrison, D. G. Pettifor and G. A. D. Briggs. ACS Nano (2009) (doi:10.1021/nn8008454)
3. J. Warner, A. A. R. Watt, L. Ge, K. Porfyrakis, T. Akachi, H. Okimoto, Y. Ito, A. Ardavan, B. Montanari, J. H. Jefferson, N. M. Harrison, H. Shinohara and G. A. D. Briggs, Nano Letters **8**, 1005 (2008)
4. J. Warner, Y. Ito, M. Zaka, L. Ge et al. Nano Letters, 8, 2328 (2008). Selected by Nature Nanotechnology as Research Highlights (doi:10.1038/nnano.2008.229).
5. J. Warner, M. Rummeli, L. Ge et al. Nature Nanotechnology 4, 500 (2009).

Acknowledgements