

# Towards Electron Spin Active Fullerene Dimers

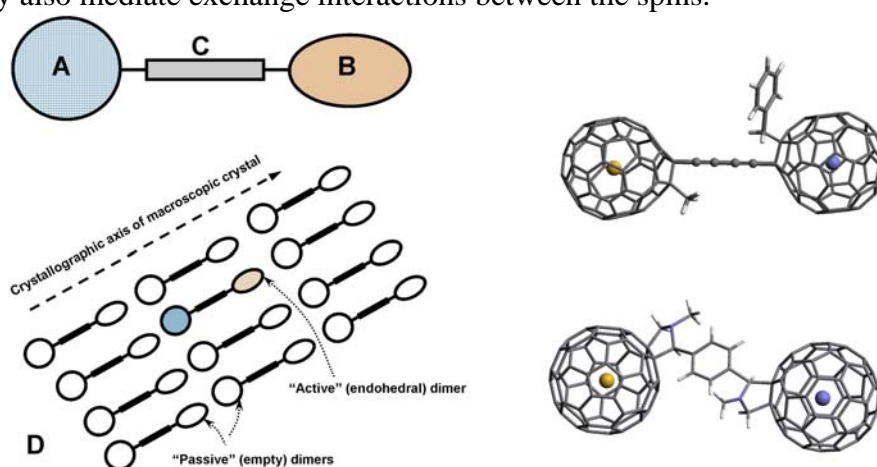
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The key aim of our collaborative project is the demonstration of entanglement between two electron spin qubits in molecular systems. Our candidate materials are fullerene dimer structures which can embody two interacting qubits such as electron spins on endohedral atoms of nitrogen incarcerated within carbon cages (i.e. N@C<sub>60</sub>). These fullerene dimers are expected to be amenable to formation of multi-qubit spin chains inside nanotubes or on 2D molecular templates. The target fullerene dimer must consist of two structurally (N@C<sub>60</sub> and N@C<sub>70</sub>) or isotopically (<sup>15</sup>N@C<sub>60</sub> or <sup>14</sup>N@C<sub>60</sub>) different parts to enable selective addressing of individual spin qubits within the dimer using pulses of microwaves (Fig. 1A/B). The two fullerene cages will be bridged by a chemical group providing a well-defined separation between the spins (Fig. 1C). The bridging group may also mediate exchange interactions between the spins.



**Figure 1.** Schematic representation of a spin-active dimer and its orientation in the matrix of non-endohedral dimers in the crystal (left). Target dimer molecules with bis-acetylene type bridge (top right), which can facilitate exchange interactions between the electron spins, and pyrrolidine type bridge (bottom right), which would allow only dipole-dipole interactions between the spins in a two-qubit fullerene system.

Various approaches for fullerene dimers using covalent functionalisation of fullerene cages have been explored in the past two or three years, however, most of them appeared to be unsuitable for endohedral fullerenes. There are three main challenges in the dimer preparation that should be addressed when designing a synthetic protocol: 1) all stages of the synthesis should be carried out under mild conditions (moderate temperature, pressure and no exposure to light); 2) chemical modifications of fullerene cages should have minimal impact on the properties of endohedral N-atom (i.e. no electronegative or highly polar groups); and 3) each step should be high-yielding and suitable for handling sub-milligram quantities of N@C<sub>60</sub>.

We have devised an efficient method for the synthesis of an asymmetric fullerene dimer which can be carried out under mild conditions (at room temperature or below) which ensures the preservation of the electron spin on the endohedral nitrogen. The fullerene cages are expected to be linked by a linear, conformationally rigid bis-acetylene spacer with delocalised  $\pi$ -electrons facilitating exchange interactions between the electron spins on endohedral N-atoms (Figure 1, top right). If this method is applicable for micro-scale synthesis, it can deliver a two-qubit molecular system required for demonstration of the key principles of quantum computing with electron spins.