AtMol deliverable reporting

**Complements for Deliverable 3D2.9:** Demonstration of a logic function with an adsorbed molecule

The deliverable 3D2.9 was set up to experimentally measure the Boolean answer of Quantum Hamiltonian Computing (QHC) molecule logic gate as designed the years before AtMol (see for example Phys. Rev. A, 78, 062316 (2008)). The uniqueness of the QHC approach is that the logical input are classical, converted in quantum information by the molecule and the logical output measured with no need of a tunnel current to pass through the complete molecule. This is an advantage compared to semi-classical intramolecular circuits. For certain quantum graphs, it was demonstrated that a QHC logic gate can performed the 6 symmetric 2 inputs – 1 output logic gates known that is the Boolean OR, AND, XOR, NXOR, NAND and NOR logic gates. A review paper about this approach was published in the Journal Advanced materials (Adv. Materials, 24, 312 (2012)) and discussed in detail during the 2nd AtMol workshop in Barcelona, January 2012.

This **Deliverable 3D2.9** was approved in advance M15 as listed in the conclusion of the 1st year AtMol Review meeting communicated to the AtMol consortium on the 30th April, 2012. The summary of the achievements of this Deliverable 3D2.9 and the works performed by P1-Toulouse, P4-Taragona and P11-Singapore are recalled in the following.

The LT-UHV-STM measurement conditions of all the gates explored by AtMol in Year 1, 2 and 3 are described in detail in the publication listed below. For example and for NOR QHC molecule logic gate, analytically pure trinaphthylene synthesized by P4-Taragona from 3-trimethylsilylnaphthyl-2-trifluoromethanesulphonate via a palladium-catalyzed [2+2+2] trimerizationwas sublimed by free evaporation in an UHV preparation chamber on a Au(111) surface kept at room temperature. Thereafter, the Au(111) sample was cooled down to 5 K. With the evaporation parameters used by P11-Singapore, coverage under 0.1 monolayer was attained. STM imaging shows molecules adsorbed on Au terraces and at step edges. P11-Singapore produced single Au atoms by a gentle crash of the W tip apex on the Au(111) surface and manipulated them one by one with the STM tip for contacting a single molecule. Up to 3 Au atoms were manipulated in close proximity to a molecule: 2 for the inputs and one as a reference to verify whether the molecule moves during one Au input operation. Standard R = 250 M low bias voltage tunnelling resistance was used for imaging and R = 0.2 M for single Au atom manipulation in pulling mode. Whenever was necessary, trinaphthylene molecule was manipulated to bring it to the Au atoms, also in pulling mode, using an R = 200 M junction resistance with a large bias voltage.

**AtMol Year 1:**

In AtMol Year 1, a coronene analogue counter was demonstrated experimentally showing that a single molecule can accommodate 3 classical inputs (see AtMol Scientific Report Year 1, Unit 3 Task 3-T2.4, Fig. 38). This was published in 2013, the time for P1-Toulouse and P11-Singapore to performed all the required ESQC STM constant current image calculations and the required PM6-CI calculations to interpret the QHC logic gate quantum behavior of the coronene molecule (Chem. Phys. Lett, 587, 35 (2013)).

Then, the experimental Boolean NOR QHC molecule logic gate response of the short Y molecule was completely probed and confirmed leading to the first ever QHC molecule logic gate (AtMol Scientific Report Year 1, Unit 3 Task 3-T2.4, Fig. 45). This was published in AtMol Year 1 (Phys. Rev. B, 155443 (2011), ACS Nano,*5* ,1436 (2011)).

**AtMol Year 2:**

After the experimental observation that a coronene molecule can take into account 3 classical inputs, P1-Toulouse explored the possibility of expending the molecule  system and demonstrated that on a nanographene like molecule, 6 classical inputs can be accommodated. (AtMol Scientific Report Year 2, Unit 2 Task 2-T1.3, Fig. 4) leading P4-Taragona to study the retro-synthesis of such a nanographene molecule.

The first XOR QHC molecule logic gate was studied experimentally in AtMol Year 2 and reported in the AtMol Scientific Report Year 2, Unit 3, Task 3-T2.4, Fig. 2. Calculations have been performed in AtMol Year 3 completed by new experiments to completely certify the XOR function before preparing the publication in 2014. The difficulties here come from the use on Al input atoms to play with the excited states of the QHC molecule logic gate.

**AtMol Year 3**

The first design of a 2 input-2output ½ adder was proposed by P1-Toulouse after a careful consideration of the QHC quantum graph theory established in AtMol Year 2 (AtMol Scientific Report Year 3, Unit 2 Task 2-T1.3, Fig. 14 & 15). It results in the retro-synthesis of the molecule by P4-Taragona and its first step of chemical synthesis (AtMol Scientific Report Year 3, Unit 2 Task 2-T2.4, Fig. 16).

Following the AtMol Year 2 number of input calculations on a Nanographene like molecule, P4-Tragagona delivered to P11-Singapore a nice nanographene molecule. On this molecule, P11-Singapore demonstrated in AtMol Year 3 that a maximum of 8 classical inputs can be experimentally accommodated by this molecule (AtMol Scientific Report Year 3, Unit 3 Task 3-T2.4, Fig. 10 & 11). This open the way to the design of more extended  systems able to perform the digital addition of 2 words of 4 digits.

**Conclusion:**

During the 3 first AtMol years, the QHC theory for molecules was proven experimentally. The NOR and XOR QHC molecule-logic gates were measured experimentally, the proof was given that on a small molecule at least 8 classical inputs can be performed experimentally after having demonstrated 3 inputs on a simple coronene molecule. The first design of a Boolean ½ adder QHC gate was proposed and the molecule chemical synthesis started. The NOR and XOR are the smallest ever experimental logic gates.