

ATOMIC PATHWAYS TOWARD THE SYNTHESIS OF FULLERENES AND TRIAZAFULLERENES FROM POLYCYCLIC AROMATIC HYDROCARBONS

*Giulio Biddau*¹, *Gonzalo Otero*², *Pablo Pou*^{1,2}, *Carlos Sánchez-Sánchez*², *Renaud Caillard*², *Celia Rogero*³, *Javier Méndez*², *Antonio. M. Echavarren*⁴, *Berta. Gómez-Lor*², *Jose Angel Martin-Gago*² and *Ruben Perez*¹

¹*Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, E-28049 Madrid, Spain*

²*Instituto de Ciencia de Materiales de Madrid – CSIC, E-28049 Madrid, Spain*

³*Centro de Astrobiología – CSIC-INTA. Ctra. de Ajalvir Km.4, 28850 -Torrejón de Ardoz, Madrid, Spain*

⁴*Institute of Chemical Research of Catalonia (ICIQ), 43007-Tarragona, Spain.*

giulio.biddau@uam.es

Resumen

True fullerene-based molecular electronics is actually limited by the current production methods. Standard methods, such as graphite vaporization, do not permit a real control on size, and, particularly, on doping (e.g. heterofullerenes and endohedral fullerene). This has promoted an intense research activity directed towards more rational and efficient synthesis methods.

We have recently achieved the formation of closed fullerenes (C₆₀) and triazafullerenes (C₅₇N₃) by thermal annealing using polycyclic Aromatic Hydrocarbons (PAHs) adsorbed on Pt(111) surfaces[1]. The PAHs (C₆₀H₃₀ and C₅₇N₃H₃₃) chosen as precursors for fullerenes and triazafullerenes (C₆₀ and C₅₇N₃), are characterized by easy synthesis and doping processes, paving the way to the formation of doped fullerene with specific characteristics.

In this communication, we focus on the theoretical aspects of that work and present large scale first principles DFT calculations using both an efficient local orbital basis[2-3] and standard plane-wave approaches [4-5]. This simulations give support for the interpretation of the experiments that confirm the feasibility of the formation process and provide insight into the atomic pathways leading from the planar PAHs to the closed fullerenes and triazafullerenes. In particular, we characterize the adsorption and STM images of both the planar precursors and the final closed molecules, considering different coverages and the influence of surface defects (like surface vacancies). Furthermore, we explore the closure process for partially- and fully-dehydrogenated precursors with the NEB method [6], identifying the relevant steps and showing that the energy barriers are low enough so they can be overcome with the available thermal energy during the annealing process.

References:

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Figures:

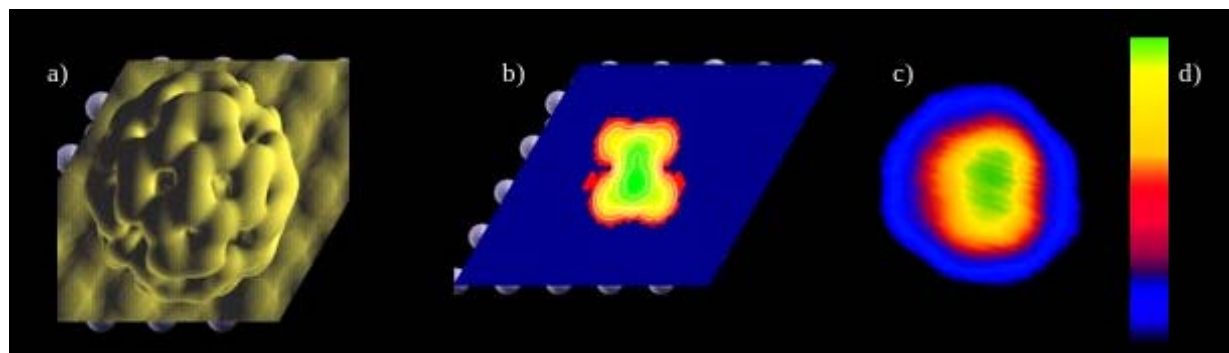


Fig.1: Adsorption of C_{60} on a dimer configuration on a Pt(111)-(5x5) unit cell. Both the charge density isosurface (a) and the charge density contours on a plane at 0.19 nm above the top carbon atoms (b) show a characteristic two-fold symmetry, with an elongated feature and a significant contribution coming from the pentagons, in agreement with the experiments (c). (d) The colour scale ranges from 0 to 0.4 nm.

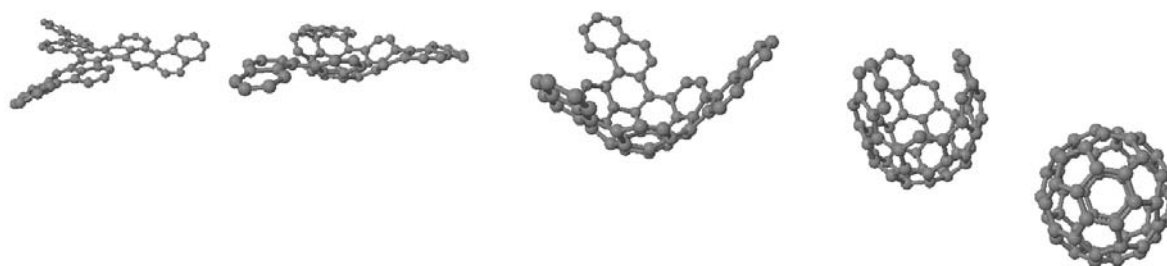


Fig.2: Closure process for the fully-dehydrogenated PAHs towards the C_{60} fullerene.