2D nanoporous graphene: a route to organic topological insulators?

The electronic and structural properties of a system are tightly connected and by experimenting with structure researchers are able to play with quantum electronic properties. Ultimately-thin materials like graphene (a single plane of carbon atoms) provide numerous illustrations of properties dominated by low-dimensionality. These properties can be enriched by engineering more advanced structures, where a "superpotential" imposed on the honeycomb lattice of graphene opens electronic band-gaps or creates new electronic bands. In particular, certain graphene-like honeycomb structures, exhibiting atomically well-defined pores regularly spaced by nanometre distances have been predicted to be two-dimensional topological insulators, *i.e.* materials that are insulators except at their edges, where the electronic structure leads to one-way conduction channels.

The family of known 2D topological insulators is restricted to only a few systems (HgTe/CdTe and InAs/GaSb heterostructures, Bismuth bilayers). A strong enough spin-orbit coupling, as conferred by the presence of high-atomic number elements, is required to open up a bandgap throughout the interior of the 2D system. The prospect for applications based on topological insulators, in the fields of spintronics, micro-electronics, metrology, and thermoelectricity, raises the key questions of their cost of production and their eco-friendliness. With these constraints in mind, our aim was to realize a 2D *organic* topological insulator based on a graphene nanoporous network.

For this purpose we used an approach exploiting molecules as building blocks (designed by F. Chérioux and co-workers at the Institut FEMTO-ST in Besançon), assembled into nanoporous networks through chemical reactions mediated by metal surfaces. Such surface-mediated chemistry is a rapidly developing field of research where only a few reaction schemes have been proposed and tested. Fig. 1 shows a Scanning Tunnelling Microscopy (STM) image of the product of a surface reaction which we have discovered: a reaction between $CO(CH_3)$ acetyl groups leaving only water as a by-product. The product is a fully covalent, atomically-thin, carbon-rich network on a metal surface, whose degree of order is currently being optimized.

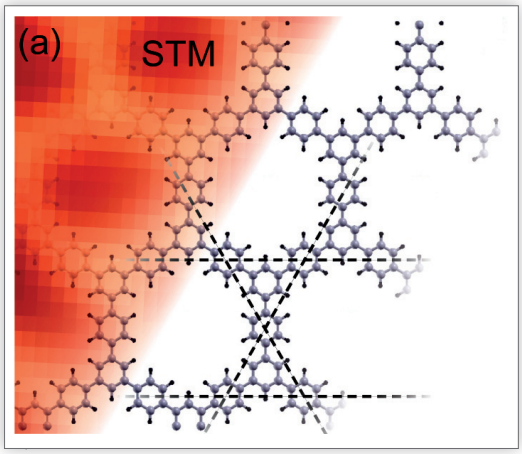

Fig. 1: *At top left, a Scanning Tunnelling Microscope image of a nanoporous network grown on a gold substrate. The superimposed schema at bottom-right shows the interpretation of this image: a hexagonal symmetry 2- Dimensional network of chains of benzene rings enclosing large "pores".*

Density functional theory (DFT) calculations were run to support the STM data and to reveal fine details of the structure that are not accessible with the spatial resolution of the STM. Also, DFT highlights the very peculiar electronic band structure of this 2D system. It shows a large band gap (larger than 2.5 eV) and a valence band characteristic of a so-called Kagome lattice – a lattice paved by hexagons each surrounded by six up – and down-pointing triangles. The calculations found one flat band, and two bands crossing at the corner of the Brillouin zone (point K in Fig. 2) with linear dispersion in the vicinity of the crossing point.

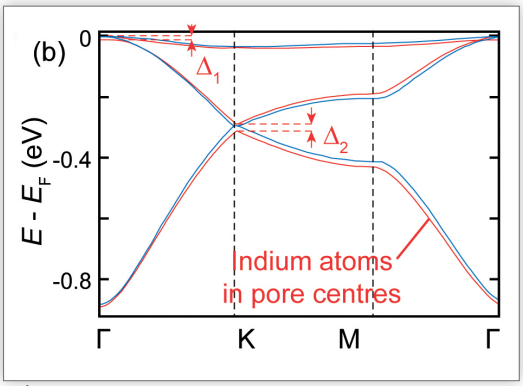

Fig. 2: *Zoom onto the valence band of the calculated band structure (electron energy vs k-vector). The blue curves are for a nanoporous network as grown in this work. Note the band degeneracies at Brouillon zone critical points K and Γ. The red curves show how placing a heavy atom (Indium, having strong spin-orbit coupling) in the pores of the lattice could remove these degeneracies (giving splittings Δ_1, Δ_2), thus producing a topological insulator.*

We also calculated the effect that adsorption, onto the lattice, of a heavy atom (Indium) with large spin-orbit coupling could have. It would open small gaps at the two degeneracy points (Γ and K in Fig. 2) turning the ideal system into an insulator. Analysis of the wave-function shows that the system indeed has non-trivial electronic properties typical of a topological insulator.

Future experimental and theoretical work will address networks formed from molecular precursors enclosing high atomic number elements which are expected to enhance the effect of spin-orbit coupling.

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FURTHER READING

"Convergent fabrication of a nanoporous two-dimensional carbon network from an aldol condensation on metal surfaces"

J. Landers, F. Chérioux, M. De Santis, N. Bendiab, S. Lamare, L. Magaud and J. Coraux

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