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"Nanoengineering"2-D nanosheets

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Top-down approaches have enabled isolation of individual 2-D nanosheets from bulk materials and have revealed extraordinary optical and electronic behaviour at the nanosheet level. An alternate potential approach to fabricate 2-D nanosheets - and materials generally - is to employ a bottom-up strategy using nanostructures as building blocks. This approach affords tunability of material properties via choice of nanobuilding blocks and material architecture, leading to a possibility of nanoengineering materials (NEMs) potentially exhibiting emergent quantum phenomena (q-NEMS).

Here, using various organic molecules and metal nanoparticles as building blocks, we present a bottom-up method to fabricate for the first time molecularly cross-linked self-assembled 2-D nanoparticle sheets (XSANS). XSANS fabricated with semiconducting "molecular wires"linkers (oligophenylene dithiol, HS-(C6H4)n-SH, with $1 \leq n \leq 3$) exhibit conductivity that decreases with increasing molecule length (the opposite trend of tunneling) and strongly enhanced molecular optical absorption (by at least ~6-orders) compared with those of unlinked molecules in solution. Spatially resolved finite difference time domain analyses and control measurements indicate that the enhancement cannot be attributed solely to strong local fields present in XSANS. However, they can be modelled provided the local complex dielectric constant is strongly modified upon cross-linking, suggesting quantum hybridization at a molecule –nanostructure (q-HYMN) level. Further, XSANS exhibit photoconductivity that reflect the hybrid molecule-metal nanoparticle character of these materials, in line with conductivity and optical results.

The present results point to XSANS, and more generally q-NEMS, as a target of opportunity for a wide range of electro-optic behaviours that can be realized from the bottom-up given the vast number of organic and inorganic nano building blocks that can be synthesized in solution.

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