Novel Graphene-Fullerene Hybrid Materials: Experiment and Theory

***Outcome*:** We have developed strategies for fabricating hybrid carbon nanostructures consisting of alternate graphene and fullerene (C_{60}) layers, which can demonstrate remarkable strength to weight ratios and tunable electronic and thermal properties.

***Impact*:** These hybrid-materials are expected to have a wide range of applications including (i) high efficiency thermoelectrics for heat harvesting, (ii) high surface area hydrogen storage media, (iii) light-weight battery-electrodes and fuel-cell membranes.

***Explanation*:** By chemically attaching alternate layers of fullerenes between successive graphene layers, mechanically robust light-weight 3-D nanostructures that harness the remarkable structure-property relations of graphene and fullerene can be synthesized, resulting in thermal and electronic properties that are tunable based on the density of fullerene interconnects between graphene layers.

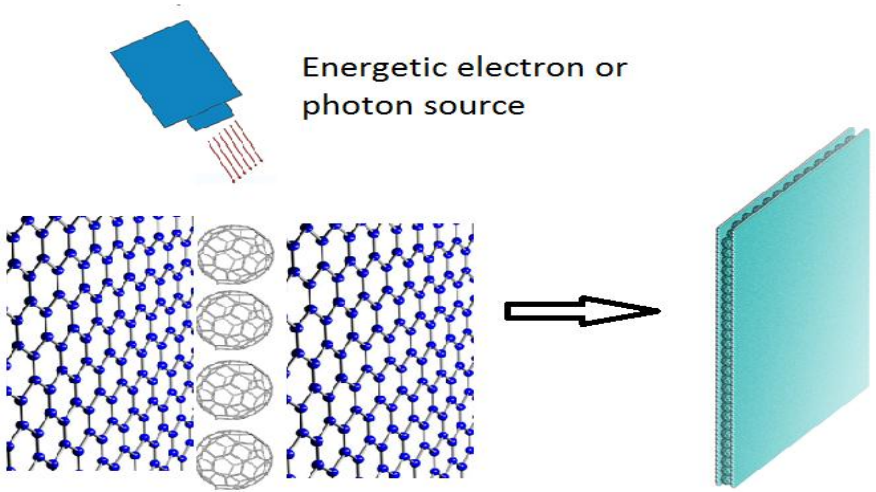

Illustration of the strategy for synthesizing graphene/fullerene/graphene building blocks

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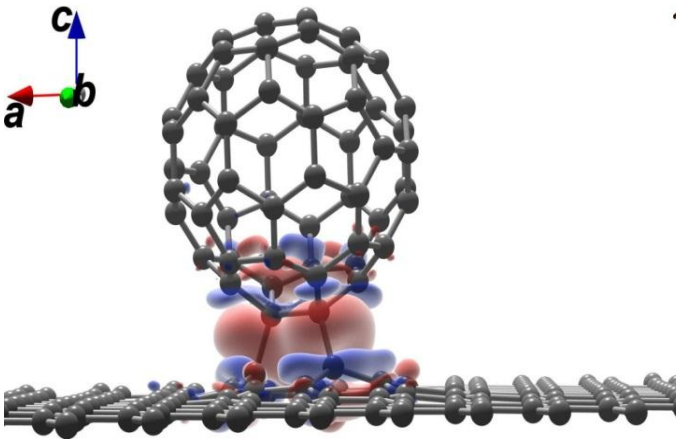
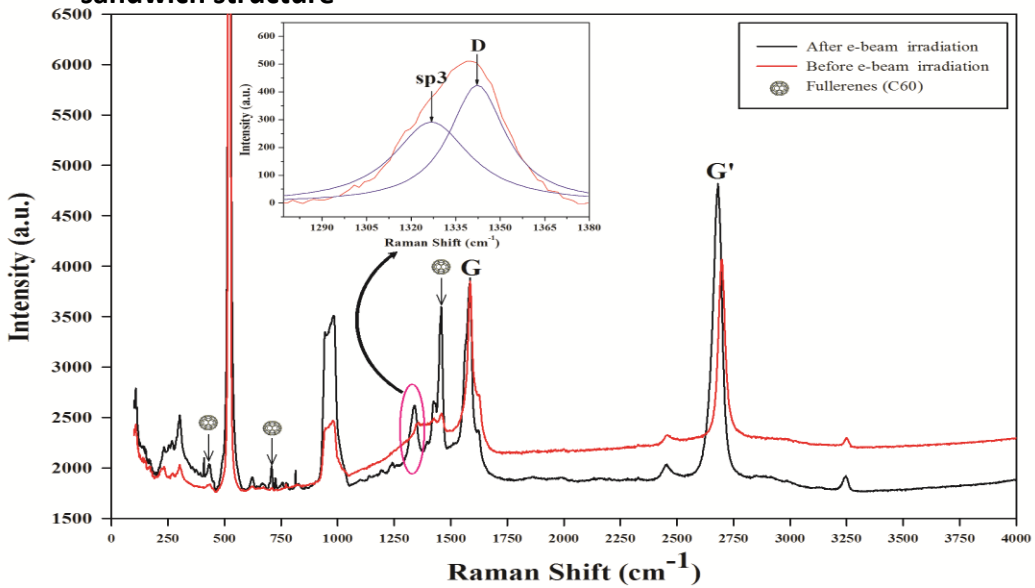
Experimental synthesis: single layer graphene (SLG) is grown by chemical vapor deposition, followed by a multilayer assembly of the SLG/C_{60}/SLG proto-mattressene structure on a silica substrate in an inert ultra-high vacuum (UHV) chamber. Electron beam irradiation of these structures stimulated **covalent** bonding between C_{60} and graphene (see Raman spectra), implying improved (i) mechanical robustness and (ii) ability to transfer energy between the graphene layers.

DFT simulations: C_{60} **covalently binds** to a graphene layer at single-vacancy defect sites, resulting in local alteration of the electronic hybridization from π–π^* to sp^3 states at the binding site (as illustrated below).

Illustration of fullerene-graphene interaction at a single-vacancy defect site modeled by DFT. The red lobes indicates electron localization and blue lobes indicate depletion, confirming the directional sp^3 bonding.



Raman spectra confirming sp^3 bonded graphene-fullerene-graphene sandwich structure



Modular Chemical-Process Center

CVD/Molecular deposition Chemical Process Center