All-carbon mixed-dimensional van der Waals heterostructures: diffusion and atomic scale deformations

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Carbon nanomaterials—including fullerenes, nanotubes and graphene—have been among the most studied structures in materials science during the past decades. Building on enormous advances in graphene research, interest has recently shifted towards the creation of so-called van der Waals heterostructures (vdWHs) [1-3]. Such materials typically combine graphene or other two- or lower dimensional molecules into vertical stacks [2-3]. The leading idea of the concept is to preserve the covalent bonding structure of the constituent molecules interacting mainly through van der Waals forces (vdW), all the while allowing strong electronic and light-matter interactions in between and within the constituent layers [4-6].

My talk will cover our recent work in fabrication and characterization of mixed-dimensional all carbon vdWHs, including buckyball sandwiches incorporating monolayers of C_{60} molecule in between two graphene sheets and single-walled carbon nanotubes (SWCNTs) likewise suspended on graphene [7,8]. In the former our room temperature scanning transmission electron microscope experiments reveal, among other things, truncated intermolecular spacing, the diffusion and rotation of individual C_{60} molecules at room temperature and finally the seizing of such motion upon merging in the electron irradiation. By acquiring several atomically resolved projections of SWCNTs on graphene the 3D shape of the interface is reconstructed through energy minimization [9], showing morphological aberrations emerging from the interlayer vdW forces. We further show that these topographic features are strain correlated but show no sensitivity to SWCNT helicity or electronic structure. Finally, we show that in an atomically clean heterostructure the competition between strain and vdW forces result in aligned carbon-carbon interfaces in some cases spanning up to several hundreds of nanometers.

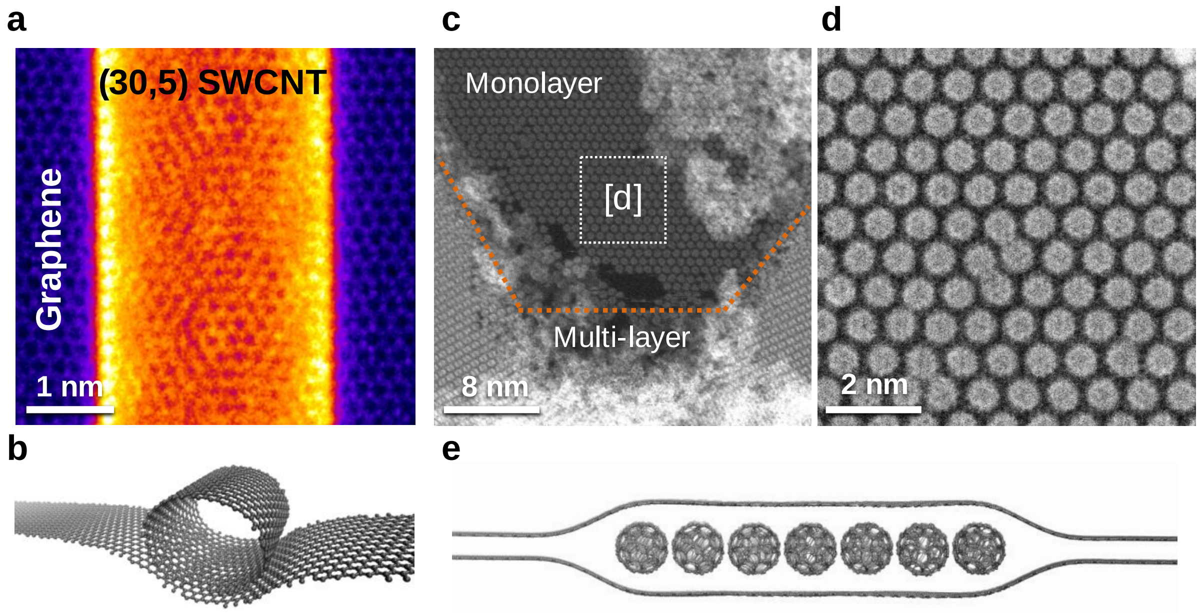

Fig. 1 **a)** A (30,5) SWCNT suspended on graphene and **b)** the recovered 3D structure of the molecular pair. **c)** An overview of a buckyball sandwich showing both mono- and multi-layer regions and **d)** an atomically resolved magnification of the monolayer and finally in **e)** a simulated sandwich structure.

[1] Geim, A. K. & Grigorieva, I. V. Van der waals heterostructures. *Nature* **499**, 419–425 (2013).
[2] Jariwala, D. *et al.* Mixed-dimensional van der waals heterostructures. *Nature materials* (2016).
[3] Liu, Y. *et al.* Van der waals heterostructures and devices. *Nature review materials* **1**, 16042 (2016).
[4] Britnell, L. *et al.* Strong light-matter interactions in heterostructures of atomically thin films. *Science* **340**, 1311–1314 (2013).
[5] Zheng, Q. *et al.* Phonon-assisted ultrafast charge transfer at van der waals heterostructure interface. *Nano letters* **17**, 6435–6442 (2017).
[6] Gjerding, M. N. *et al.* Layered van der waals crystals with hyperbolic light dispersion. *Nature communications* **8**, 320 (2017).
[7] Mirzayev, R. *et al.* Buckyball sandwiches. *Science Advances* **3**, e1700176 (2017).

[8] Mustonen, K. *et al.* atomic-scale deformations at the interface of a mixed-dimensional van der Waals heterostructure , Submitted.
[9] Hofer, C. *et al.* Revealing the 3D structure of graphene defects, Submitted.