

Graphene hybrids and extended defects: Revealing 3D structures, dynamics, and new insights to radiation damage

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Obtaining the location of every atom in three dimensions is arguably the ultimate goal of electron microscopy. Electron tomography is usually based on a large number of projections, and requires that the sample does not change its structure during data acquisition. Here, we demonstrate that the location of every atom in graphene defects and individual and clustered silicon impurities in graphene can be obtained from only two projections [1,2]. Importantly, two projections could be obtained with no changes in the atomic network, while larger numbers of exposures (e.g. for a complete tomographic series) would be prohibited by the limited radiation stability of the sample. The analysis reveals a kink in the graphene sheet at grain boundaries that depends on the misorientation angle. For 3-coordinated silicon impurities, we could confirm the out-of-plane position of the Si atom from direct images and also follow beam-induced dynamics where the impurity changes from one side of the graphene sheet to the other, in both directions. The 3D structure could also be obtained for a graphene sheet that interacts with cleanly deposited single-walled carbon nanotubes [3]. In this hybrid system, the van der Waals interaction leads to a deformation of both the carbon nanotube and the graphene sheet. Finally, we have studied hybrids of graphene and molecular structures, namely C60 fullerenes [4] and chlorinated copper phthalocyanine (ClCuPc) [5]. They represent the first cases that mono-layers of organic molecules were studied by electron microscopy at atomic resolution. In the graphene-fullerene sandwich, dynamics of entire molecules can be observed with weakly bound fullerenes oscillating between different positions at the edges of 2D C60 molecular crystals. Moreover, we observed the transition from rotating individual fullerenes through dimers with suppressed rotation to molecular clusters locked into position. Atomic resolution studies of ClCuPc monolayers on graphene provided new insights to radiation damage. The critical dose of the mono-layers is orders of magnitude lower than that of bulk crystals, and the molecular fragments form cross-linked networks on the graphene surface after the loss of the chlorine atoms.

