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Reversible intermediate energy state of CVD graphene: A signature of rippling?

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The application of graphene in device technologies will require that the electronic band-structure of different graphene materials is measured in detail and that graphene-substrate interactions are well understood. Both, the degree of sp²-hybridisation and the electronic band-structure can be directly probed with NEXAFS. Furthermore the technique enables detailed studies of structural changes at the graphene surface and at its substrate interface.

Our NEXAFS studies at the Australian Synchrotron have produced new evidence for a contentious state in graphene near 288 eV. This resonance has been intermittently observed before by others and it is often referred to as an 'interlayer state' due to a perceived analogy with graphite.

For CVD-graphene synthesized on copper we see a pronounced anisotropy for this state and derive an excitation energy of 288.3 eV and a partial overlap with an isotropic contaminating resonance. After annealing and keeping the graphene in ultra-high vacuum, the NEXAFS signature of the 288.3 eV state only gradually appears and builds-up over several hours. This signature can be removed again by renewed annealing. The reversible phenomenon may thus relate to residual lattice mismatch between the graphene and the copper substrate. Associated stress may gradually be relaxed through the rippling of the graphene layer. Tilting angles of >20° appear possible. The rippling is evidenced in our data by a correlated, reversible non-linearity of the cos-square-theta-dependence of the 285 eV π^* resonance of graphene.

Keywords

Graphene, NEXAFS, interface, CVD, surface topology, soft x-rays

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