In this proposal, the spectroscopic investigation of functionalized mono– and few– layered graphene (FLG) has been performed. The samples were made by the proposer as highly crystalline layers grown by precipitation on SiC and by chemical vapour deposition on Ni(111) surfaces. Their electronic, vibronic and optical properties have been spectroscopically investigated by a combined experimental and theoretical approach. In low dimensional and strongly anisotropic systems, correlation effects play a crucial role in understanding and describing the electronic and vibronic properties. Therefore the particular focus of this proposal lies on correlation effects, i.e. the renormalization of the non–interacting electron and phonon dispersion relations of FLG and doped graphene layers. These correlation effects comprise electron–electron, electron–phonon and electron–plasmon coupling. These are the underlying processes that are key for unravelling (1) transport, (2) vibronic and (3) optical properties in FLG, GICs and related structures such as nanoribbons, nanotubes and fullerenes.

We have investigated the distinct changes of the electronic band structure upon (1) covalent, (2) substitutional and (3) ionic functionalization. To that end we have performed functionalization of graphene on metals by (1) hydrogen, (2) nitrogen and (3) potassium. It has been shown that the relevant physics in each case is quite different. The huge charge transfer of alkali metals results in an increase of the Fermi level, and the lattice distortion induced by hydrogenation causes defect scattering and bandgap opening. Regarding substitutional doping by nitrogen impurities, it was shown that sp2 bonded nitrogen transfers charge to graphene.

We have also found a new electronic state in H-graphene that is located between the π and π * bands. For undoped H-graphene this state is energetically situated within the gap around EF and is accessible with absorption spectroscopies such as NEXAFS. In the case of n-doped H-graphene the midgap state becomes available for electrons and directly observable with ARPES since it is then situated below EF. Therefore, the H impurity band likely acts as an electron acceptor level which provides the possibility to control the electron concentration in H-graphene via the H/C ratio. An estimation of the Mott criterion and a calculation of the typical DOS suggests that above H/C ~ 1% and below H/C ~ 6%, the acceptor level can form an extended impurity band

The list below depicts the publications that have been achieved for this project.

10.1103/PhysRevB.80.085423 Phonon surface mapping of graphite: Disentangling quasi-degenerate phonon dispersions A. Grüneis Physical Review B - Condensed Matter and Materials Physics 8 / 80 American Physical Society

01/08/2009

Yes 2 10.1103/PhysRevB.86.155151 Experimental and computational insight into the properties of the lattice-mismatched structures: Monolayers of h-BN and graphene on Ir(111) D. Usachov Physical Review B - Condensed Matter and Materials Physics 15 / 86 American Physical Society

01/10/2012

Yes 3 10.1088/0953-8984/25/4/043001 Synthesis and electronic properties of chemically functionalized graphene on metal surfaces Alexander Grüneis Journal of Physics Condensed Matter 4 / 25 Institute of Physics Publishing

30/01/2013 043001 -

Yes

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4 10.1103/PhysRevB.83.165433 Direct observation of a dispersionless impurity band in hydrogenated graphene D. Haberer Physical Review B - Condensed Matter and Materials Physics 16 / 83 American Physical Society

01/04/2011

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Yes 5 10.1103/PhysRevB.82.075415 Quasifreestanding single-layer hexagonal boron nitride as a substrate for graphene synthesis D. Usachov Physical Review B - Condensed Matter and Materials Physics 7 / 82 American Physical Society

01/08/2010

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Yes 6 10.1021/nl101066m Tunable Band Gap in Hydrogenated Quasi-Free-Standing Graphene D. Haberer Nano Letters 9 / 10 American Chemical Society

08/09/2010 3360 - 3366 10.1021/nl101066m Yes