Doping graphene up to and beyond the van Hove singularity by rare earth intercalation

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The intercalation of graphene on SiC(0001) is a versatile route to decouple graphene from the substrate and simultaneously tune its electronic properties. The electron doping of graphene can be steered by the intercalation of different atomic species. Theoretical work predicts correlated electron states such as charge or spin density waves and chiral superconductivity (SC) when the charge carrier concentration in graphene is high enough in the vicinity of the van Hove singularity (VHS) [1].

High electron doping can be realized by intercalation of rare earth elements like Gd [2], Yb [3,4] and Tb [5], which leads to a flat band scenario (extended VHS) exhibiting a high density of states along the $\overline{KMK'}$ direction of the Brillouin zone (BZ), as observed by angle-resolved photoelectron spectroscopy (ARPES). In the case of Yb the doping can be steered around the VHS: by diluting the amount of Yb underneath the graphene layer the VHS is moved into the unoccupied states [3], while additional adsorption of potassium on top of the graphene leads to a shift of the flat band below the Fermi level leading to a so-called overdoping of graphene [4]. Furthermore, hybridization of the graphene π^*-bands with the Yb 4f electron states leads to a strong renormalization of the band structure of graphene [3].

Intercalation of Tb leads to a doping beyond the VHS with the flat band residing 70 meV below the Fermi level (Fig 1). Based on the random phase approximation, we could show that the graphene hosts a SC state with a critical temperature in the regime of 300 to 600 mK [5]. A spectral function analysis of the π^*-bands in the Eliashberg framework indicates an electron-phonon coupling constant of ~0.49, which is below a theoretically estimated critical value of 0.56, indicative of a stable d-wave SC, in contrast to a conventional phonon-driven s-wave SC state.

Besides the strong doping phase, which does not show any additional diffraction spots in low-energy electron diffraction (LEED), we observed a new superstructure by LEED when depositing a higher amount of Tb and flash annealing to temperatures around 1200 °C. Our photoemission studies hint towards the formation of a Tb-carbide phase by reaction with the carbon atoms of the graphene layer.

References:
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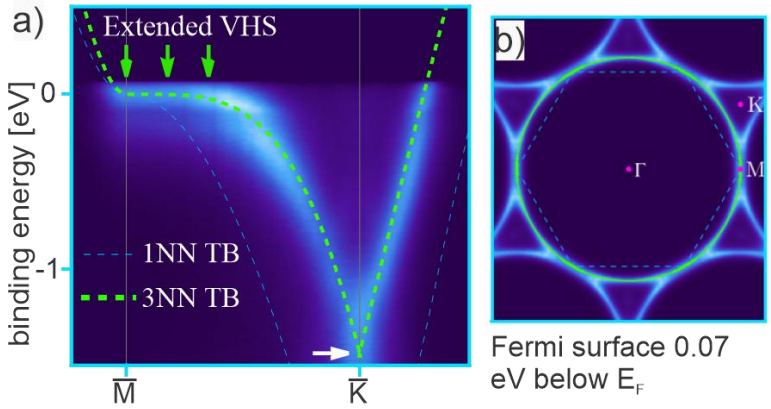

Fig. 1: Band structure of Tb intercalated graphene: (a) Extended VHS (green arrows) measured along the $\overline{MKΓ}$-direction. The Dirac point (white arrow) is at approx. −1.55 eV. The graphene band dispersion is well reproduced by the overlaid 3rd nearest neighbor tight binding (3NN TB) model (dashed green lines) while a 1st NN TB model (dashed blue lines) does not capture the experimental band structure. (b) Constant-energy surface measured at the VHS, 0.07 eV below the Fermi level; some high-symmetry points of the (1 × 1) BZ are indicated. (Adopted from Ref. [5]).