Calcium intercalation of epitaxial graphene

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Realizing proximity-induced correlation effects, such as superconductivity, in graphene is a major challenge in the field of research on two-dimensional (2D) materials. Among the graphite intercalation compounds (GICs), the Ca-GICs exhibit the highest critical temperature with T_c=11.5 K, where the electron doping from the Ca atoms may be a crucial factor.

Hence, expanding this superconducting phase into the 2D limit of epitaxial monolayer graphene (EMLG) on SiC(0001) requires precise control over the arrangement of the intercalated Ca atoms. However, several structural models have been proposed for Ca-intercalated EMLG, and in particular, the vertical position - i.e., the gallery - in which Ca resides remains unresolved [1,2].

Here, we investigate Ca-intercalation into EMLG and zero-layer graphene (ZLG) grown by polymer-assisted sublimation growth. Using x-ray and angle-resolved photoemission spectroscopy, we monitor the intercalation process and quantify the doping level. Our results indicate the saturation of the SiC interface by Ca. We observe the low-energy bands of the resulting intercalated bilayer graphene (see Figure 1, left), and more interestingly regarding the superconducting state, a highly doped graphene π-band (Figure 1, right), which may point towards the presence of calcium in the upper gallery. In the case of ZLG, Ca intercalation leads to decoupling from the substrate and, consequently, to the formation of a graphene π band. We also observe strong electron doping for this band, comparable to the result obtained for the EMLG system. Based on these results, this contribution will discuss possible scenarios for the structural model of this system.

References:
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[2] J. C. Kotsakidis et al., Chem. Mater. 32 (2020) 6464.

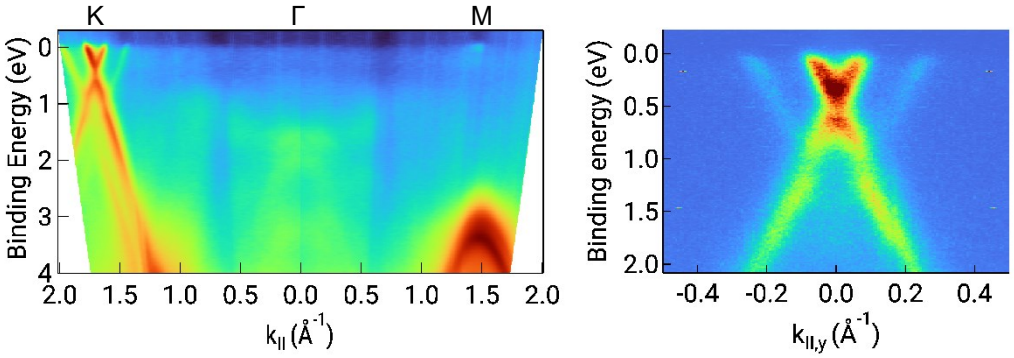

Fig. 1: ARPES map of Ca-intercalated EMLG along the ΓK and ΓM high symmetry directions of graphene (left) and in the vicinity of the graphene K point (right).