Charge transfer and enhanced metallicity via alkali metal doping on Nanoporous Graphene

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The modification of graphene (Gr) electronic and vibrational properties via electron doping, is a topic of great interest both for applications and fundamental physics, ranging from energy storage [1], to the emergence of exotic states like electron-phonon induced superconductivity [2]. Alkali metal (AM) adsorption, represent a stable and controlled way to dope Gr without affecting its planar configuration, thus suitable to investigate the electronic and vibrational response of enhanced metallicity in Gr. Herewith, we present AM doping on Gr via the adsorption of different alkali metals (Na, K, Cs), on a novel fully freestanding nanoporous graphene (NPG, fig. 1a), a three-dimensional architecture with electronic and vibrational hallmarks of an ideal suspended graphene [2]. In this study, we investigated the effects of the electron doping combining different spectroscopic techniques in ultra-high vacuum environment. We followed the evolution of the core levels and the spectral density of states at the Fermi level by photoemission (fig.1 b-c): the electron migration leads to the occupation of the π^* -states, yielding a rigid shift of the bands with the formation of a charge carrier dependent π^* Dirac plasmon; the latter is also directly probed via electron energy loss spectroscopy [3]. The increased metallicity is then correlated with the non-adiabatic vibrational response of the system, showing strong modification of the Raman bands, as a function of the charge transfer and electron-phonon coupling. Our findings unveil a distinct π^* -states occupation and π^* - Dirac plasmon evolution as a function of the different alkali metal and charge carrier transfer.

References

- [1] Zhang et I., J. Mater. Chem. A, 2021, 9, 27140.
- [2] Profeta et al., Nature Phys, 2012, 8, 131–134.
- [3] Di Bernardo et al., ACS Omega 2017, 2, 7, 3691–3697.
- [4] Marchiani et al., Nano Lett. 2023, 23, 1, 170–176.

Figures

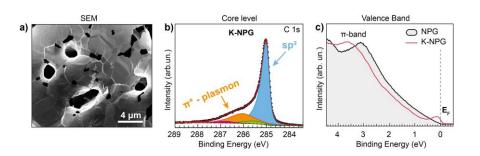


Figure 1: a) Scanning electron microscopy (SEM) image of an NPG sample. b) C 1s core level deconvolution of a highly doped K-NPG showing the typical main sp² component and the doping induced π^* -plasmon loss. c) Valence band spectral density of states of the NPG and K-NPG at saturation, displaying a rigid shift of the band as the consequence of electronic occupation.

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