

Dual path hydrogenation of the graphene/Ni(111) interface.

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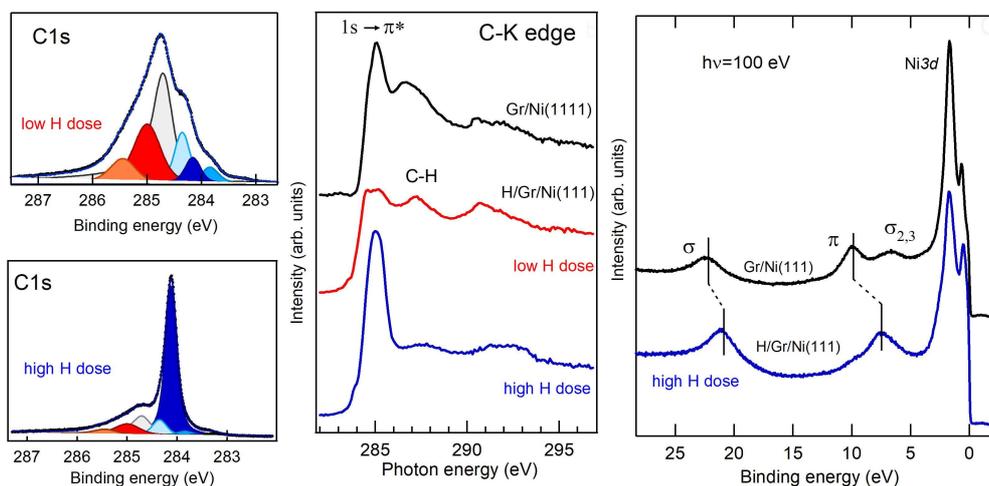
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The control of hydrogen adsorption on graphene and the thermodynamic stability of hydrogenated graphene are fundamental topics in view of new potentialities and possible future developments of nanostructured carbon-based materials in the field of hydrogen storage.

Hydrogenation of epitaxial graphene supported on transition metal crystals, such as Ir(111) and Pt(111), has been observed to proceed through the chemisorption of H atoms, whose lateral distribution at the nanoscale can be driven by the corrugated moiré structure of graphene. Our results demonstrate that a more complex scenario arises when graphene is supported on Ni(111).

The room temperature hydrogenation of Gr/Ni(111) by means of a flux of H atoms has been investigated by combining X-ray photoelectron (XPS) and absorption (XAS) spectroscopies, temperature programmed desorption (TPD) and density functional theory (DFT) calculation. We found that at first H atoms chemisorb on the graphene surface up to a coverage well below 0.5 monolayers. The formation of C-H bonds determines new components in the C1s core level spectrum that DFT calculations attribute to C atoms directly bonded to H and to their first neighbours. As the H exposure is prolonged beyond the saturation of the chemisorbed phase, a slow but continuous H intercalation occurs. Valence band and NEXAFS measurements demonstrate that the H-intercalated graphene is completely decoupled from the Ni substrate. TPD measurements show that the intercalated H atoms desorb at lower temperature with respect to those bonded directly to graphene and indicates the penetration of H atoms in the metal bulk.



(left) C1s core level spectra measured on the Gr/Ni(111) surface exposed to low (top) and high (bottom) H doses and corresponding to the chemisorbed and intercalated phases, respectively; (middle) XAS and (right) valence band spectra measured at different hydrogenation stages.