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Combining nitrogen substitutional defects and oxygen intercalation to control the graphene corrugation and doping level

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Abstract

By means of Scanning Tunneling Microscopy (STM) experiments and first-principles calculations, we demonstrate the synergetic effect of the combination of two different strategies to modify the properties of graphene supported on a strongly interacting substrate like Rh. A complete control of the corrugation and doping level is achieved combining the introduction of nitrogen defects and oxygen intercalation. Firstly, we show how to use ion bombardment to obtain purely-substitutional N-doped graphene on Rh(111) with tunable dopant concentration. In a second step, the interaction with the substrate is controlled by the amount of intercalated oxygen atoms. Unlike weakly interacting substrates, the highly corrugated structure of G/Rh(111) leads to remarkable variations of the electronic properties associated with nitrogen defects created in the high and low areas of the moiré. After oxygen intercalation, the N-doped graphene layer decouples from the substrate preserving the incorporated nitrogen atoms, which display a subtle dependence of the STM contrast. First–principles calculations confirm the identification of substitutional N-defects and the recovery of the Dirac cone with a tunable shift governed by the nitrogen concentration. Our results support the combination of different modification techniques to tailor structural and electronic properties of graphene and other 2D materials.

Keywords: Graphene, Nitrogen doping, Scanning Tunneling Microscopy, Density Functional Theory

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