

Raman Spectroscopy of Intercalated Epitaxial Graphene on SiC(0001)

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Raman spectroscopy provides a powerful and non-destructive probe of graphene, offering rich information on its structural, electronic, and vibrational properties, including strain, doping, and electron-phonon interactions.[1] In this work, we employ resonance Raman spectroscopy to investigate epitaxial graphene on SiC(0001) achieved by intercalation with different species, including hydrogen (H), indium (In), and tin (Sn) into the zeroth layer of graphene (ZLG), as well as the as-grown monolayer graphene (MLG) system.

Epitaxial graphene on SiC offers a scalable and stable platform, while intercalation enables precise control over the graphene-substrate interaction, allowing both the decoupling of graphene into quasi-free-standing layers and the formation of novel interfacial phases beneath graphene.[2,3] Through systematic Raman analysis, we examine the evolution of strain and charge doping across these systems, revealing distinct signatures associated with each intercalant and interface configuration. In particular, we demonstrate the formation of a metallic triangular Sn(1×1) phase beneath graphene, which, together with an on-surface plasmonic Sn nanoantenna, gives rise to enhanced light-matter interaction via gap-mode plasmonic effects.[4] This results in more than two orders of magnitude enhancement of the graphene Raman response, accompanied by modified selection rules and intrinsic electron-phonon coupling.[5] Furthermore, we investigate the thermomechanical behavior of these systems and show that the properties of the graphene layer are strongly governed by the nature of the interfacial coupling.

Our results establish Raman spectroscopy as a versatile tool to characterize intercalated epitaxial graphene systems and provide insight into the interplay between graphene and engineered buried interfaces, with implications for tunable electronic, plasmonic, and optoelectronic applications.

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