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Ionic permeability and interfacial doping of graphene on SiO_2

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Abstract

Thanks to its outstanding electrical properties and chemical stability, graphene finds widespread use in various electrochemical applications. Although the presence of electrolytes is known to strongly affect its electrical conductivity, the underlying mechanism has remained elusive. Employing terahertz spectroscopy as a contact-free means, we investigate the impact of ubiquitous cations (Li^+, Na^+, K^+, Ca^{2+}) in chloride-based aqueous solution on the electronic properties of SiO_2-supported, p-doped graphene. We find that cations can induce a positive shift in Fermi energy of 200 meV in graphene, with the kinetics of that shift and the final doping level being determined by cation size and concentration. Combined with theoretical calculations, we show that this ionic doping effect in graphene involves cationic permeation through defects in graphene. The interfacial cations at the graphene-SiO_2 interface electrostatically eliminate the substrate doping effect in graphene. These insights are crucial for electrochemical applications including energy storage, and ionic sensing.

Figures

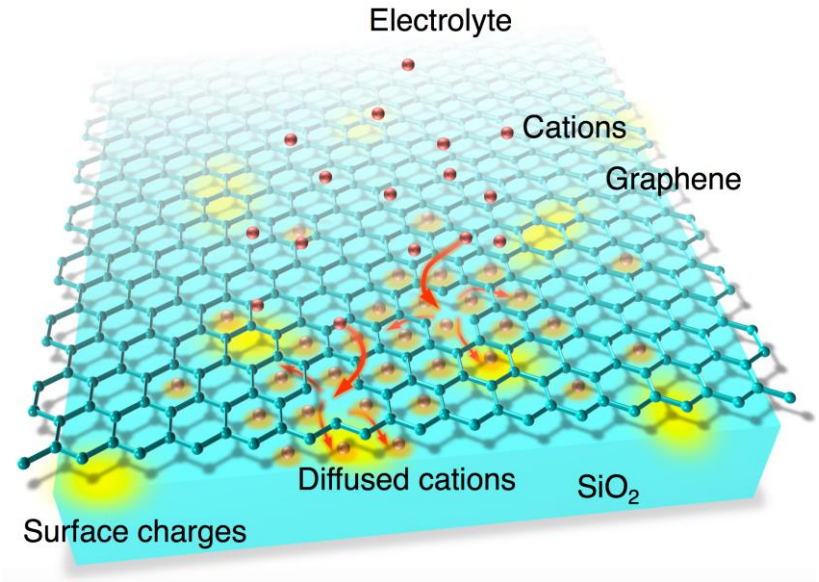

Figure 1: Illustration of cation permeation through graphene sheet to the graphene-SiO_2 interface which results in the interfacial doping of graphene.