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ABSTRACT

The Graphene-Liquid-Interface investigated by Electrochemical Surface Plasmon Resonance

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Single sheets of graphene are highly promising as active elements of electrical and electrochemical sensors, [1] since the material is just surface. Being just a single layer of carbon atoms, they are ideally suited as *metal-free* electrodes for studying the behavior of (electro)catalyst particles in operando / in situ. Understanding the physicochemical properties and mechanistic details of phenomena occurring at the graphene-liquid-interface (GLI) is fundamentally important for an optimal use of the electrodes and sensors in a desired application. In our group, on the one hand, we are interested in developing analytical methods to gain an improved understanding of the GLI. On the other, the unique properties of the GLI are exploited to design novel sensors.

This talk will focus on the use of electrochemical surface plasmon resonance (EC-SPR) detection [2] for the study of interfacial phenomena and processes at the GLI. Typically, in SPR sensing, the plasmonic properties of a gold film are used to sense interfacial interactions. Using graphene directly on a sensor chip has several disadvantages due to the strong interaction between the gold surface and the graphene sheet. Here we present two new methods to overcome this disadvantage. The first one involves the use of a van der Waals heterostructure comprised of graphene and a hexagonal boron nitride (hBN) sheet to decouple the graphene sheet from the underlying gold film. This enables us to monitor electrochemical phenomena on the graphene surface using the plasmonic signals. [3] For example, this provides the capability to distinguish if the interfacial electron transfer reaction occurs by diffusion or adsorption of the redox active species. In a second method, we have realized a polymer layer as a dielectric plasmon waveguide on an SPR sensor, followed by transfer of a graphene sheet electrode on top. Using such an architecture, we show that the electrochemical nucleation and dissolution kinetics of metal nanoparticles can be followed in situ on graphene. Taken together, EC-SPR constitutes a complementary and versatile tool to study interfacial processes at the GLI.

Literature

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[3] R. Jungnickel, K. Balasubramanian, chemRxiv 2024, DOI: 10.26434/chemrxiv-2024-z82f7