Modifying 2D Material Interfaces through Chemistry and Controlled Stacking for Engineering Applications

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Our lab at MIT has been interested in how 2D electronic materials such as graphene can be utilized to advance new material concepts. Of particular interest to us has been how such materials participate in electron transfer chemistries as in the case of graphene spontaneously reacting with benzene diazonium salts, as well as electrochemical modification of 2D materials. This presentation will outline our work in applying and modifying electron transfer theory to extend to 2D materials, explicitly accounting for the electronic contributions due to the substrate. We show a stark difference in the rate of electron-transfer reactions with organic diazonium salts for monolayer graphene supported on a variety of substrates. Reactions proceed rapidly for graphene supported on SiO_2 and sapphire, but negligibly on alkyl-terminated and hexagonal boron nitride (hBN) surfaces, as shown by Raman spectroscopy. We also develop a model of reactivity based on substrate-induced electron-hole puddles ingraphene, and achieve spatial patterning of chemical reactions in graphene by patterning the substrate. Field-effect transistor (FET) devices composed of a MoS_2- graphene heterostructure can combine the advantages of high carrier mobility in graphene with the permanent band gap of MoS_2 for digital applications. We also investigate the electron transfer, photoluminescence, and gate-controlled carrier transport in such a heterostructure. We show that the junction is a Schottky barrier, whose height can be artificially controlled by gating or doping graphene. Atomically thin MoS_2 is of great interest for electronic and optoelectronic applications because of its unique two-dimensional (2D) quantum confinement, however, the scaling of optoelectric properties of MoS_2 and its metallic junctions with layer number remains unaddressed. We utilize photocurrent spectral atomic force microscopy (PCS-AFM) to image the current and photocurrent generated between a biased PtIr tip and MoS_2 between n = 1 to 10 layers. Dark current measurements in both forward and reverse bias reveal characteristic diode behavior well described by Fowler-Nordheim tunneling with a monolayer barrier energy of 0.605 eV and an effective barrier scaling linearly with layer number. Under illumination at 600 nm, the photocurrent response shows a marked decrease up to n = 4 but increasing thereafter, which we describe using a model that accounts for the linear barrier increase at low n, but increased light absorption at larger layer number creating a minimum at n = 4. Comparative 2D Fourier analysis of physical height and photocurrent images shows high frequency spatial variations in substrate/MoS_2 contact that exceed the frequencies imposed by the ITO substrates. These results should aid in the design and understanding of optoelectronic devices based on quantum confined MoS_2. Lastly, we highlight emerging applications of 2D material materials.