PhD THESIS DEFENSE: Mid-Infrared Surface Sensing Based on Two-Dimensional Materials

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Mid-infrared (mid-IR) spectroscopy in the wavelength region between 2 and 20 ?m is a powerful technique to identify vibrational absorption signatures of molecules, finding in this way extensive applications in healthcare, environmental monitoring, and chemical analysis. Enhanced IR light-molecules interactions can be achieved by exploiting nanostructured surfaces supporting polaritons - hybrid excitations of light and dipolar elements of matter. Recently, polaritons of two-dimensional van der Waals (2D-vdW) materials unveiled a vibrant playground for mid-IR spectroscopy as they possess remarkable properties such as light trapping at deep nanoscale. This dissertation aims to investigate 2D-vdW materials for technological sensing applications. Hence, we explore the mid-IR sensing performance of nanostructures of widely studied 2D-vdW crystals: graphene (the pioneering vdW material

with tunable plasmon polaritons) and hexagonal boron nitride (hBN, sustaining ultralow-loss phonon polaritons). Relevant functionalization layers, such as polymer adsorber and antibodies, are combined with the 2D-vdW nanostructures to create gas and for bio-molecular sensors, respectively.
Here, we present three main experimental works of 2D-vdW-based mid-IR molecular sensing. First, we investigate the CO2 detection using graphene nanoribbons functionalized with ultrathin CO2-chemisorbing polyethylenimine (PEI). The localized surface plasmon resonance (LSPR) of graphene is modulated by varying CO2 gas concentration, whose substantial shifts are influenced by the reversible PEI-induced doping of graphene. Second, we examine the phonon-enhanced CO2 detection of hBN nanoresonators functionalized with thin PEI layer. The phonon-polariton resonance is modulated by varying CO2 levels with high signal-to-noise ratio signals. Third, we present a quantitative bioassay by transducing different vitamin B12 target concentrations into LSPR shifts of bio-functionalized graphene nanostructures (subsequent addition of pyrene linkers and recombinant anti-vB12 antibody fragments). Additionally, we observed the same result-trends for the same bioassay using graphene nanostructures fabricated both by small-scale (i.e., electron beam lithography) and large-scale (i.e., nanoimprint lithography) methods.
Our proof-of-concept mid-IR sensing experiments show quantitative results for the detection of gas and biomarker with functionalized 2D-vdW nanostructures. The opportunity of combining the mid-IR spectroscopy with industrially large-scale 2D-vdW nanostructures (e.g., nanoimprinted GNH in this dissertation) would enable cost-effective technologies in future developments. This dissertation contributes to the field of 2D-vdW-based mid-IR spectroscopic sensors towards exploring novel designs and improved sensitivity, which eventually could lower the limit of detection for molecular analytes in various applications.

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