Low-frequency modes, twisting- and defect-induced shifts in Raman modes in MoS_2, $MoSe_2$, and phosphorene

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Abstract

Beyond graphene materials, such as transition metal dichalcogenides (TMDs) and black phosphorus (or "phosphorene" at the monolayer limit) have attracted significant attention as emerging 2D materials due to their unique properties compared with well-explored graphene. [1] Compared to graphene, these materials present band gaps that can offer much in terms of potential electronic applications. Many characterization techniques have been employed to improve the understanding of these materials, to establish their crystal structure, purity, number of layers, and internal arrangements (i.e., relative orientation of stacked layers). As a non-destructive and fast technique, Raman has repeatedly proven effective for quantitative determination of these properties. However, many details of the experimental measurements cannot be directly understood from the intuition developed with the vast experience with graphene. Instead, the interpretation of many experimental features requires a dedicated modeling effort based on first-principles methodologies. Fortunately, at the same time as experimental characterization and sample preparation techniques have evolved to new heights, theoretical schemes are now combined with unprecedented computational resources to provide tools akin to a *virtual microscope* to enable the translation of experimental data into fundamental understanding of intrinsic properties of the investigated samples. [2] Here, I will summarize how density functional theory and non-resonant Raman scattering methodologies are combined to address many issues that are central to the scientific and technological development of TMDs, phosphorene, and other materials. In particular, I will discuss the importance of low-frequency modes in the study of layer-layer interactions in TMDs [3] and phosphorene [4], how relative twisting angles can be determined by monitoring relative shifts in Raman active mode in $MoSe_2$ [5], and how Raman can be employed to understand in-plane anisotropy in phosphorene [6,7]. Finally, I will show how defect concentration (notably, S vacancy density) can be determined by the sole knowledge of the shift in major vibration modes of MoS_2 subjected to electron irradiation [8].

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