Observation of Interlayer Phonons in Transition Metal Dichalcogenide Atomic Layers and Heterostructures

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Interlayer phonon modes in atomically thin transition metal dichalcogenide (TMD) heterostructures were observed for the first time. We measured the low-frequency Raman response of MoS$_2$/WSe$_2$ and MoSe$_2$/MoS$_2$ heterobilayers. We discovered a distinctive Raman mode (30 - 35 cm$^{-1}$) that cannot be found in any individual monolayers (see Fig. 1). By comparing with Raman spectra of bilayer (2L) MoS$_2$, 2L MoSe$_2$ and 2L WSe$_2$, we identified the new Raman mode as the layer breathing mode (LBM) arising from the perpendicular vibration between the two TMD layers. The LBM only emerges in bilayer regions with atomically close layer-layer proximity and clean interface. In addition, the LBM frequency exhibits noticeable dependence on the relative orientation between the two TMD layers, which implies a change of interlayer separation and interlayer coupling strength with the layer stacking.

We also investigated the ultralow-frequency Raman response of atomically thin ReS$_2$, a special type of TMD with unique distorted 1T structure. We found that the two shear modes in bilayer ReS$_2$ are nondegenerate and clearly resolved in the Raman spectrum (see Fig. 2), in contrast to the doubly degenerate shear modes in other two-dimensional materials. By carrying out comprehensive first-principles calculations, we can account for the frequency and Raman intensity of the interlayer modes and determine the stacking order in bilayer ReS$_2$. Few-layer ReS$_2$ exhibits rich Raman peaks at frequencies below 50 cm$^{-1}$, where a panoply of interlayer shear and breathing modes are observed.