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Observation of Interlayer Phonons in Transition Metal Dichalcogenide Heterostructures RUI HE, ZHIPENG YE, CHAO JI, CASIE MEANS-SHIVELY, HEIDI ANDERSON, TIM KIDD, University of Northern Iowa, KUANG-CHANG CHIU, CHENG-TSE CHOU, JENN-MING WU, YI-HSIEN LEE, National Tsing Hua Univ, Taiwan, TROND ANDERSEN, MIT, CHUN HUNG LUI, UC Riverside — Interlayer phonon modes in transition metal dichalcogenide (TMD) heterostructures are observed for the first time. We measured the low-frequency Raman response of MoS₂/WSe₂ and MoSe₂/MoS₂ heterobilayers. We discovered a distinct Raman mode (30 - 35 cm⁻¹) that cannot be found in any individual monolayers. By comparing with Raman spectra of Bernal bilayer (2L) MoS₂, 2L MoSe₂ and 2L WSe₂, we identified the new Raman mode as the layer breathing vibration arising from the vertical displacement of the two TMD layers. The layer breathing mode (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 rotational angle between the two TMD layers, which implies a change of interlayer separation and interlayer coupling strength with the layer stacking.

Rui He
University of Northern Iowa

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