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Spatially Resolved Raman Spectroscopy of Single- and Few-Layered WS₂¹ AYSE BERKDEMIR, The Pennsylvania State University, HUMBERTO R. GUTIERREZ, University of Louisville, ANDRES R. BOTELLO-MENDEZ, Universite Catholique de Louvain, NESTOR PEREA-LOPEZ, ANA L. ELÍAS, CHENG-ING CHIA, BEI WANG, VINCENT H. CRESPI, FLORENTINO LOPEZ-URIAS, The Pennsylvania State University, JEAN-CHRISTOPHE CHARLIER, Universite Catholique de Louvain, HUMBERTO TERRONES, MAURICIO TERRONES, The Pennsylvania State University — We systematically investigated the Raman scattering of single- and few-layered WS₂ as a function of the number of S-W-S layers and the excitation laser wavelength in the visible range (488, 514 and 647 nm). For the three excitation wavelengths used in this study, the frequency of the A_{1g}(Γ) phonon mode monotonically decreases with the number of layers, while the E_{2g}¹(Γ) frequency increases. For single-layer WS₂, 514.5 nm excitation generates a second-order Raman resonance for the longitudinal acoustic mode at the M point. This 2LA(M) resonance results from a double-resonant Raman coupling between the electronic band structure and lattice vibrations, an effect not previously seen in any single-layered metal dichalcogenide. We performed ab initio calculations to determine the electronic and phonon band structures of single-layer and bulk WS₂, these results were used to compute the reduced intensity of the 2LA mode from the fourth order Fermi golden rule. Our observations establish an unambiguous and nondestructive Raman fingerprint for identifying single- and few-layered WS₂ islands.

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