

Electronic Supplementary Information

Phonon and Raman scattering of two-dimensional transition metal dichalcogenides from monolayer, multilayer to bulk material

Xin Zhang, Xiao-Fen Qiao, Wei Shi, Jiang-Bin Wu, De-Sheng Jiang, and Ping-Heng Tan

State Key Laboratory of Superlattices and Microstructures,
Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China

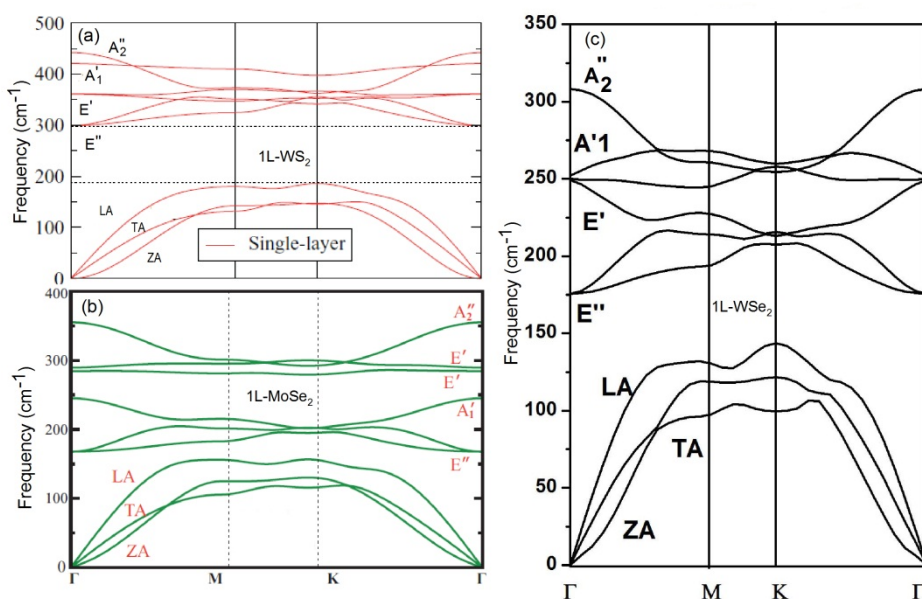


Fig.S1 Phonon dispersion of (a) 1L-Ws₂, (b) 1L-MoSe₂ and (c) 1L-WSe₂. Reproduced with permission from ref.1. Copyright 2011, American Physical Society. Reproduced with permission from ref.2. Copyright 2013, American Physical Society. Reproduced with permission from ref.3. Copyright 2014, Nature Publishing Group.

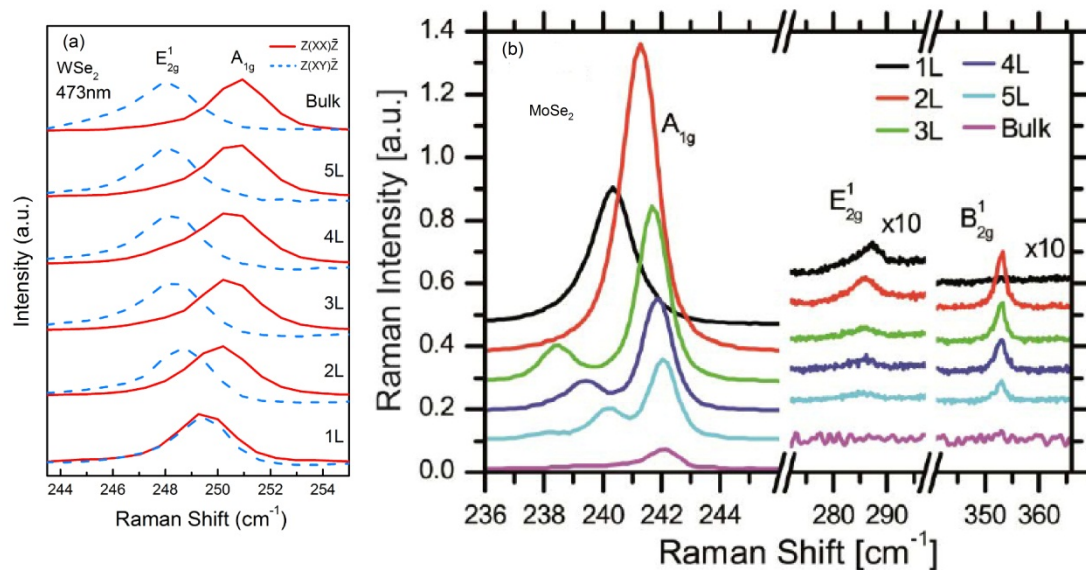


Fig.S2 (a) Polarized Raman spectra of 1-5L and bulk WSe₂ excited by 473nm. Here, E_{2g}¹ mode increases in frequency while A_{1g} mode decreases when reduced from bulk to monolayer. Two modes become almost degenerate in 1L-WSe₂, which is identical to the calculated phonon dispersion in Fig.S1. Reproduced with permission from ref.4. Copyright 2013, Royal Society of Chemistry. (b) Raman spectra of 1-5L and bulk MoSe₂. Decreasing in frequency of A_{1g} is obvious when thinned down to monolayer, while E_{2g}¹ is much weak and its increasing is clearly revealed in 1-3L. Reproduced with permission from ref.5. Copyright 2013, The Optical Society.

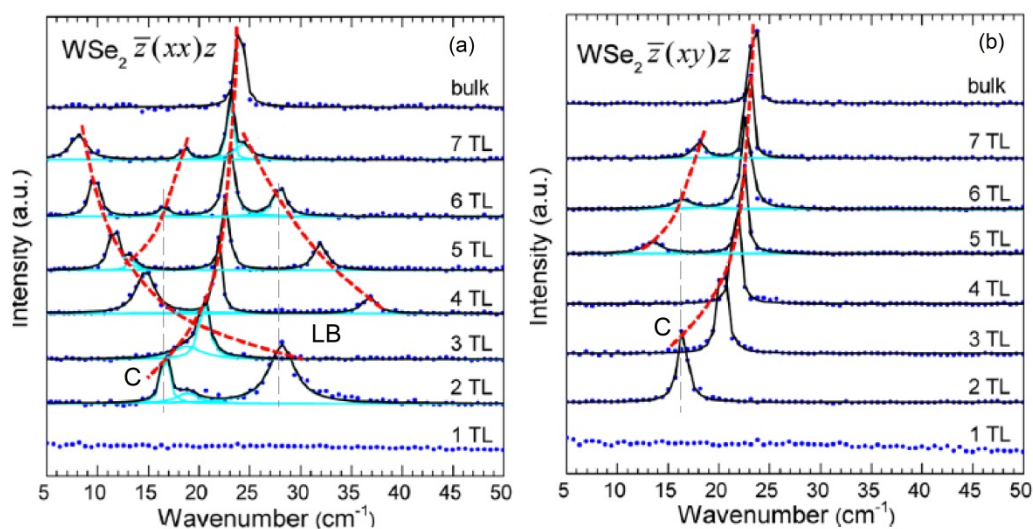


Fig.S3 Polarized ULF Raman spectra of 1-7L and bulk WSe₂ under (a) $\bar{z}(xx)z$ and (b) $\bar{z}(xy)z$ configurations. Red dashed lines are from the original paper (ref. [6]). Two gray dashed lines denote the C and LB modes from 2L. According to Fig. 6 in main text, C and LB modes from 2L stay almost unchanged with increasing layer thickness. C and LB modes with similar frequency are expected in 2nL-WSe₂ (n=1, 2, 3...). The frequency branches can be reproduced based on Fig. 6. Reproduced with permission from ref.6. Copyright 2013, American Chemical Society.

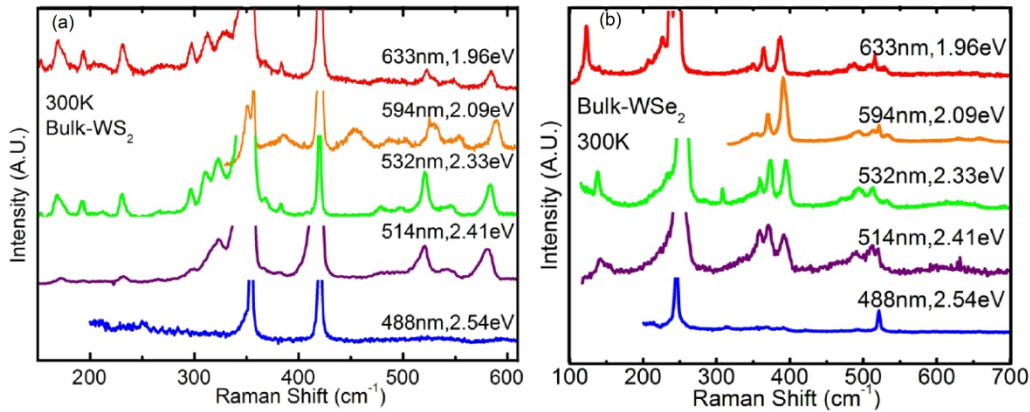


Fig.S4 Raman spectra of bulk (a) 2H-WS₂ and (b) 2H-WSe₂ at five excitation wavelengths. Reproduced with permission from ref.7. Copyright 2014, AIP Publishing LLC.

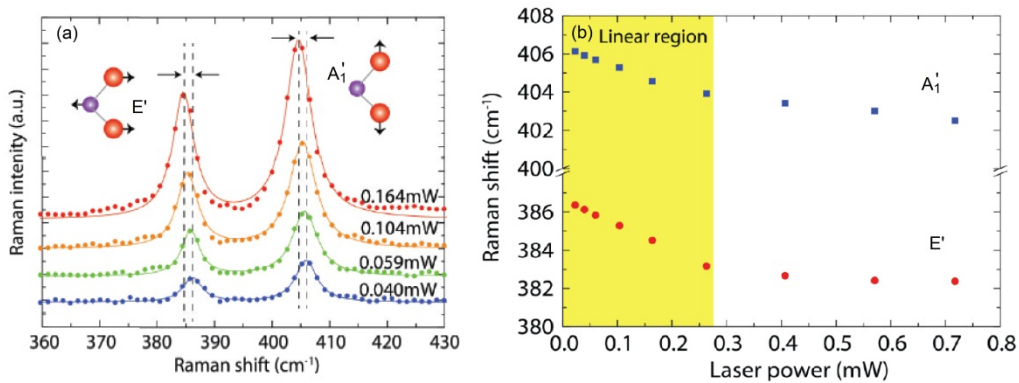


Fig.S5 (a) Four example Raman spectra of suspended, monolayer MoS₂ at increasing excitation laser power at Room Temperature in air environment. (b) The Raman peak frequencies of E' and A₁' modes for laser power up to 0.72 mW for the suspended monolayer MoS₂ flake. Nonlinearity in the Raman peak position occurs above approximately 0.3 mW, beyond the linear region shown in yellow. Reproduced with permission from ref.8. Copyright 2014, American Chemical Society

Reference

- [1] A. Molina-Sanchez and L. Wirtz, Phys. Rev. B **84** (2011).
- [2] H. Terrones, E. Del Corro, S. Feng, J. M. Pomirol, D. Rhodes, D. Smirnov, N. R. Pradhan, Z. Lin, M. A. T. Nguyen, A. L. Elias, et al., Sci. Rep. **4**, 4215 (2014).
- [3] S. Horzum, H. Sahin, S. Cahangirov, P. Cudazzo, A. Rubio, T. Serin, and F. M. Peeters, Phys. Rev. B **87**, 125415 (2013).
- [4] W. Zhao, Z. Ghorannevis, K. K. Amara, J. R. Pang, M. Toh, X. Zhang, C. Kloc, P. H. Tan, and G. Eda, Nanoscale **5**, 9677 (2013).
- [5] P. Tonndorf, R. Schmidt, P. Bottger, X. Zhang, J. Bönner, A. Liebig, M. Albrecht, C. Kloc, O. Gordan, D. R. T. Zahn, et al., Opt. Express **21**, 4908 (2013).
- [6] Y. Y. Zhao, X. Luo, H. Li, J. Zhang, P. T. Araujo, C. K. Gan, J. Wu, H. Zhang, S. Y. Quek, M. S. Dresselhaus, et al., Nano Lett. **13**, 1007 (2013).
- [7] J. H. Fan, P. Gao, A. M. Zhang, B. R. Zhu, H. L. Zeng, X. D. Cui, R. He, and Q. M. Zhang, J. Appl. Phys. **115**, 053527 (2014).
- [8] R. Yan, J. R. Simpson, S. Bertolazzi, J. Brivio, M. Watson, X. Wu, A. Kis, T. Luo, A. R. Hight Walker, and H. G. Xing,

