Using Low-Frequency Raman Spectroscopy to Probe the Interlayer Stacking Configurations of MoS_2/WS_2 Heterobilayers

Hui-Yu Cheng[1*], Wei-Ting Hsu[1], Po-Hsun Wu[1], Li-Syuan Lu[1], Yu-Ming Chang[2], and Wen-Hao Chang[1]

[1]*Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan*

[2]*Center for Condensed Matter Sciences, National Taiwan University, Taipei, Taiwan*

* Email:cabin610082@gmail.com

Abstract

Heterostructures of transition metal dichalcogenides (TMDs) have emerged as new building blocks for developing novel electronic and optoelectronic devices. For heterobilayers formed by vertical stacking of different monolayer TMDs, the electronic and optical properties depend strongly on the stacking configuration due to the symmetry-dependent interlayer coupling. It has been established that the stacking configuration of TMD homobilayers can be characterized by low-frequency (< 50 cm^{-1}) shear and breathing Raman modes, corresponding to in-plane and out-of-plane layer vibrations, respectively. Here we show that the low-frequency Raman modes can also be used to identify the stacking configuration of MoS_2/WS_2 heterobilayers. We observed a systematic difference in Raman frequencies for heterobilayers with parallel (AA) and antiparallel (AB) stacking, consistent with theoretical predictions. The Raman tensors of the low-frequency interlayer vibration modes are further investigated by polarization-resolved measurements. We demonstrate that probing the low-frequency interlayer Raman modes is a powerful and noninvasive way for identifying the stacking configuration of TMD heterobilayers

WS_2/MoS_2 Heterobilayers

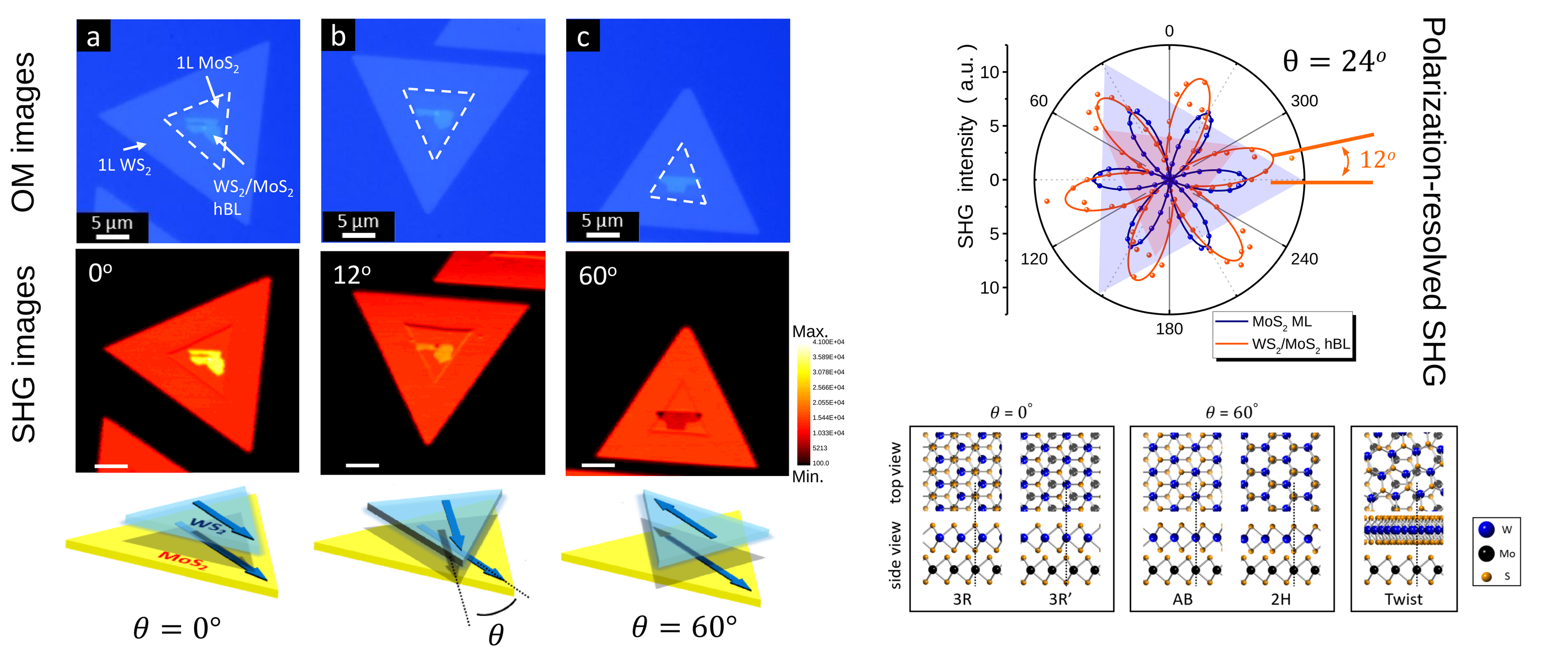

Fig. 1. Optical microscope and polarization-resolved SHG measurement can identify the stacking configuration and interlayer twist angles, but not the structures which had the same twist angle.

Experimental Setup

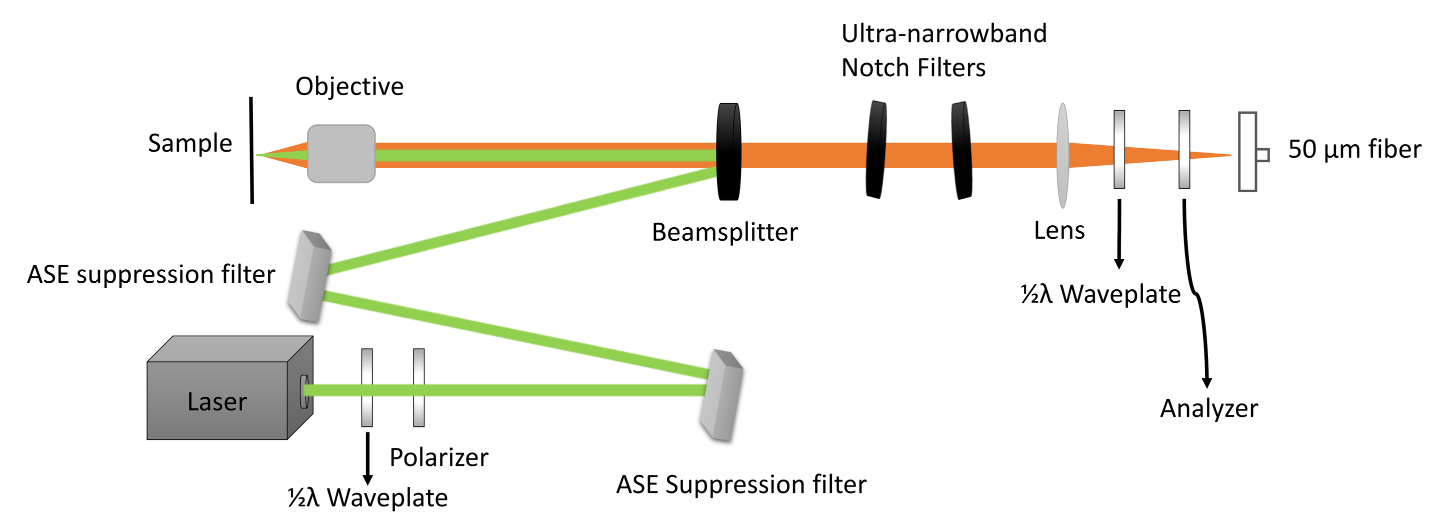

Fig. 2. All the experiments were performed on a home-built confocal spectromicroscopy using a 100x NA 0.9 objective, providing ~0.3 μm spatial resolution. Suppression filters suppress the broad spectrum of spontaneous emission that commonly occurs in laser diodes to provide a pure, single frequency laser. Ultra-narrowband notch filters with laser line attenuation of up to 99.9% and transition width of <10 cm^{-1} can dramatically improve the ability of the Raman system to resolve low frequency Raman scattering. This region of spectrum reveals a new "Structural Fingerprint" to complement the traditional "Chemical Fingerprint" of Raman.

THz Raman Spectra of WS_2/MoS_2 Heterobilayers

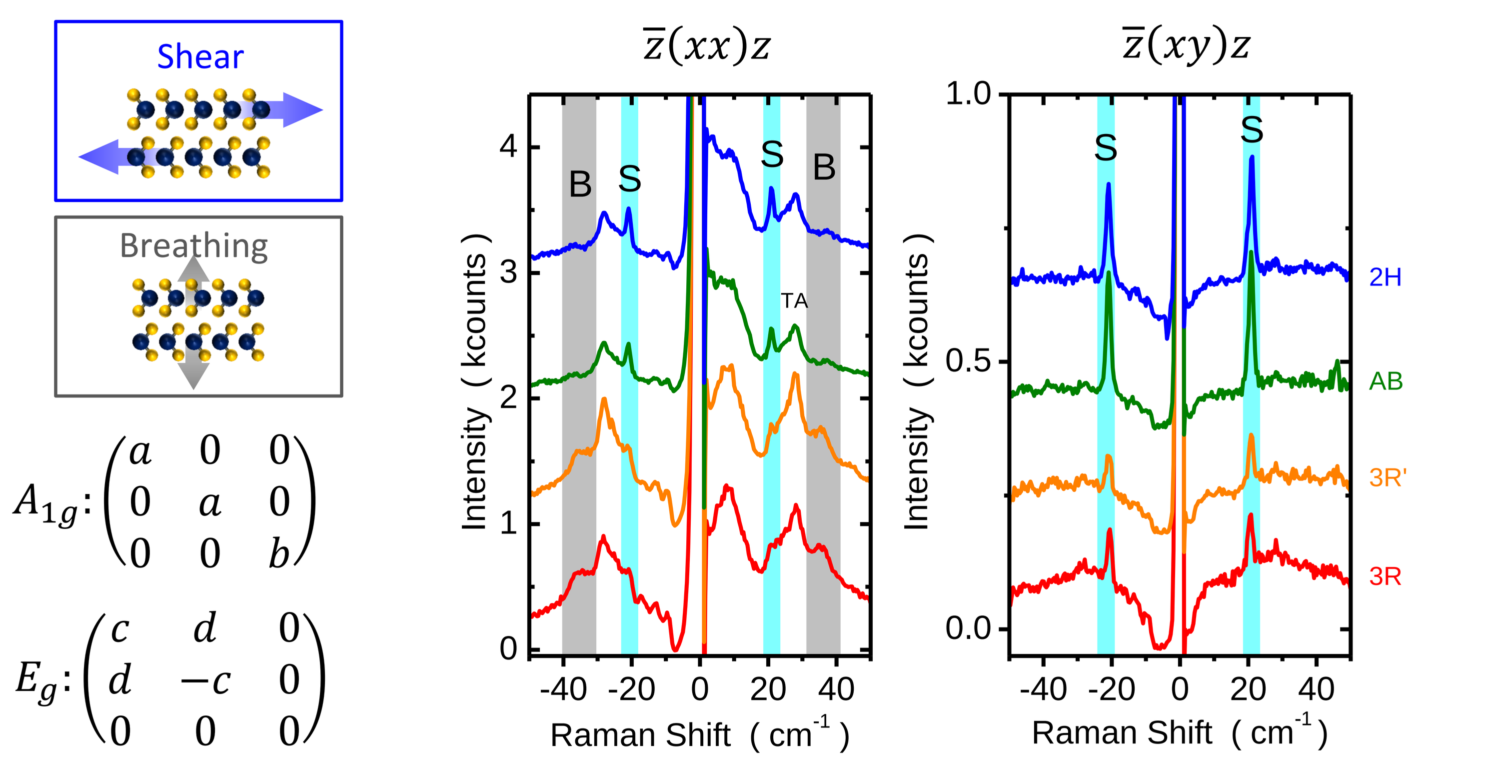

Fig. 3. Raman spectra of WS_2/MoS_2 heterobilayers with different twisting angles selected 0° and 60° under parallel (VV) and cross (VH) polarization configurations. The breathing modes A_{1g} can only be observed under the $\bar{z}(xx)z$ polarization; the shear modes E_g can be observed under both $\bar{z}(xx)z$ and $\bar{z}(xy)z$ polarization. This is helpful for the analysis of the shear mode phonon peaks. To investigate the interlayer vibrations, we determine the peak position to a precision of ~0.1 cm^{-1} by curve-fitting.

Statistical Histogram

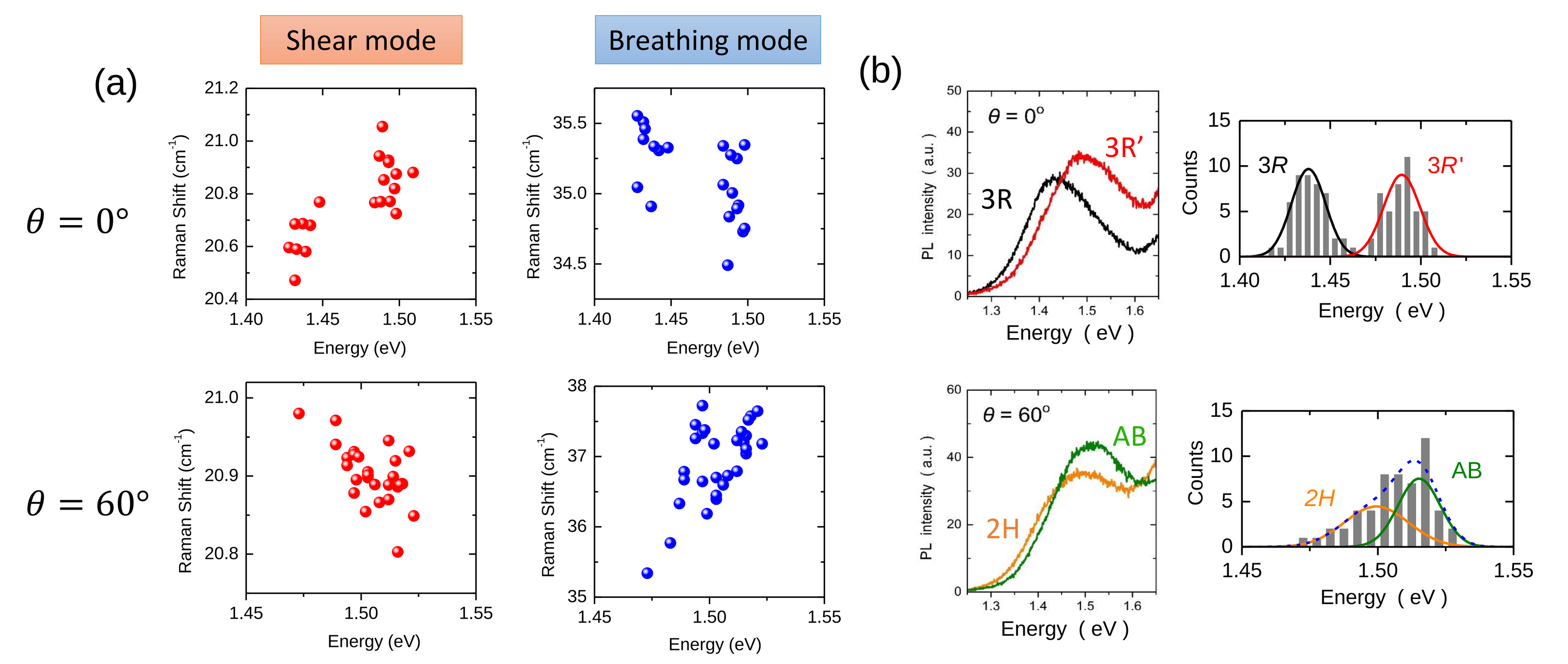

Fig. 4. (a) Statistical analysis of Raman spectroscopic measurements. (b) Statistical histogram of PL peak energy of WS_2/MoS_2 heterobilayers used to differentiate the crystal structure.

Summary for Interlayer Phonon Modes

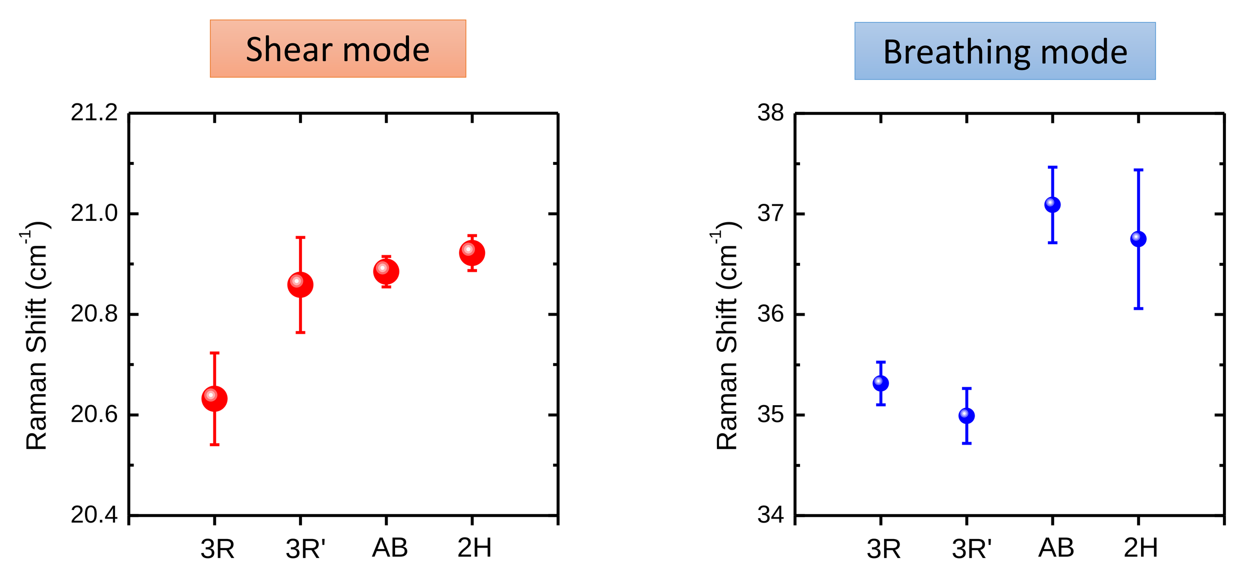

Fig. 5. Summary for Interlayer shear and breathing modes.

THz Raman Spectra of Twisted Heterobilayers

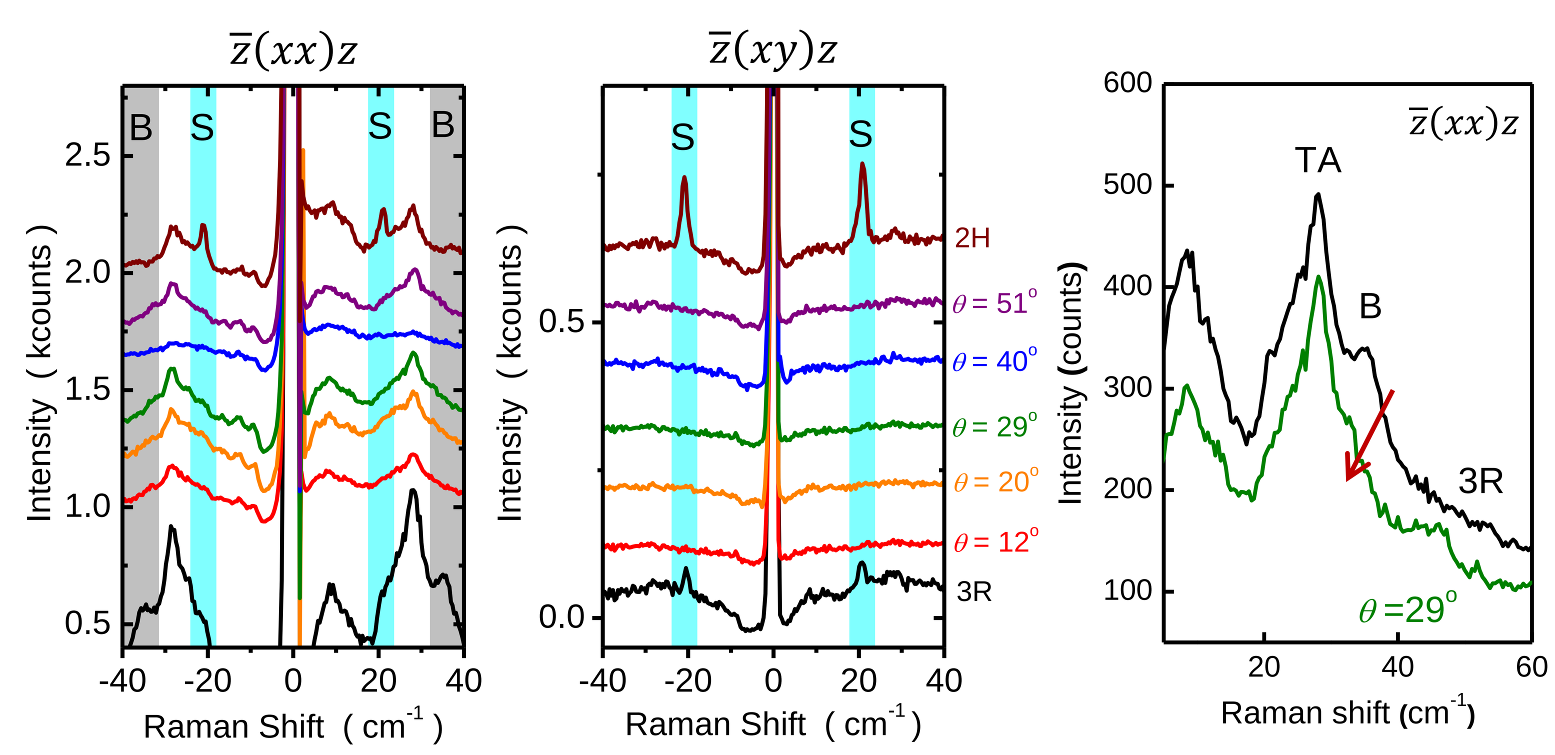

Fig. 6 Raman spectra of twisted WS_2/MoS_2 heterobilayers. The low frequency interlayer breathing mode for twist angles other than 0° and 60° shows a shift (red arrow) toward lower wavenumber.

Conclusion

✓ The twisted WS_2/MoS_2 heterobilayers with parallel (AA) and antiparallel (AB) can be distinguished by breathing and shear modes under parallel (VV) and cross (VH) polarization configurations.