Kangwon Kim
Sanghun Kim, Jae-Ung Lee, Hyeonsik Cheong
Department of Physics, Sogang University, Seoul 04107, Korea
hcheong@sogang.ac.kr

Excitonic Resonance Effects and Davydov Splitting in Circularly Polarized Raman Spectra of Few-layer WSe_2

We studied excitonic resonance effects of few-layer tungsten diselenide (WSe_2) by using circularly polarized Raman spectroscopy with up to eight excitation energies. The main E_{2g}^1 and A_{1g} modes of few-layer WSe_2 near 250 cm^{-1} appear as a single peak in the Raman spectrum taken without consideration of polarization but are resolved by using circularly polarized Raman scattering [1]. There are two resonance behaviors of the E_{2g}^1 and A_{1g} modes: firstly, both the E_{2g}^1 and A_{1g} modes are enhanced near resonances with the exciton states. Secondly, the A_{1g} mode exhibits Davydov splitting for trilayers or thicker near some of the exciton resonances. In addition, we observed a Breit-Wigner-Fano type signal at ~301 cm^{-1} only for 1.58-eV excitation, and its origin is found to be the interplay between two-phonon scattering and indirect band transition. The low-frequency Raman spectra show shear and breathing modes involving rigid vibrations of the layers and also exhibit strong dependence on the excitation energy. An unidentified peak at ~19 cm^{-1} that does not depend on the number of layers appears near resonance with the B exciton state at 1.96 eV (632.8 nm). The strengths of the intra- and inter-layer interactions are estimated by comparing the mode frequencies and Davydov splitting with the linear chain model [2, 3], and the contribution of the next-nearest-neighbor interaction to the inter-layer interaction turns out to be about 34% of the nearest-neighbor interaction [4].

References

[1] S. Y. Chen *et al.*, Nano Lett., 15 (2015) 2526.
[2] G. Froehlicher *et al.*, Nano Lett., 15 (2015) 6481.
[3] K. Kim *et al.*, ACS nano, 10 (2016) 8113.
[4] S. Kim *et al. submitted.*

Figures

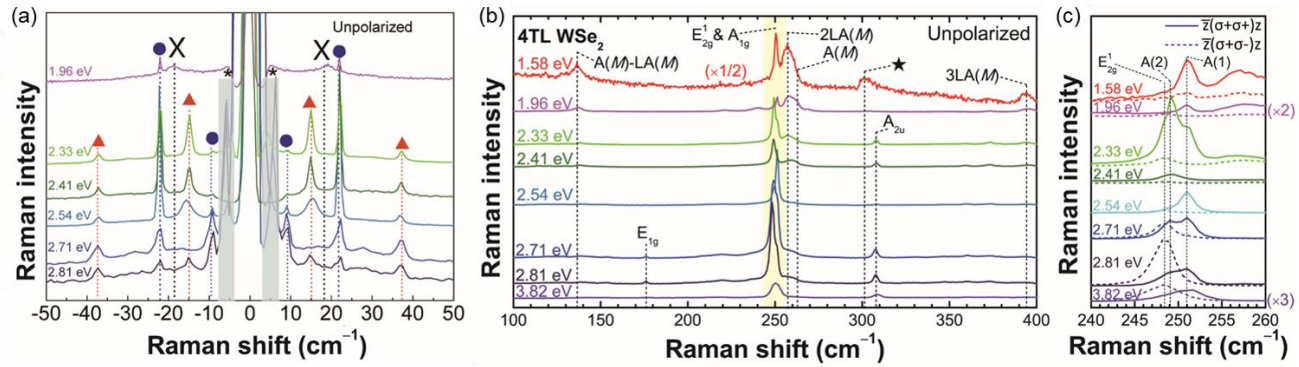

Figure 1: Raman spectra of 4L WSe_2 by using several excitation energies (a) in low-frequency reign and (b) in high-frequency reign. (c) Circularly polarized Raman spectra of 4L WSe_2 by using eight excitation energies.