

Supporting Information for Deciphering the Intense Postgap Absorptions of Monolayer Transition Metal Dichalcogenides

Jinhua Hong^{*†}, Masanori Koshino[†], Ryosuke Senga[†], Thomas Pichler[‡], Hua Xu[§], Kazu Suenaga^{*†,¶}

[†]Nanomaterials Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8565, Japan

[‡]Faculty of Physics, University of Vienna, Strudlhofgasse 4, A-1090 Vienna, Austria

[§]Key Laboratory of Applied Surface and Colloid Chemistry, School of Materials Science and Engineering, Shaanxi Normal University, Xi'an 710119, P. R. China

[¶]The Institute of Scientific and Industrial Research (ISIR-SANKEN), Osaka University, Ibaraki 567-0047, Japan

*Email: jinhuahong436@gmail.com (J. H.), or suenaga-kazu@sanken.osaka-u.ac.jp (K. S.)

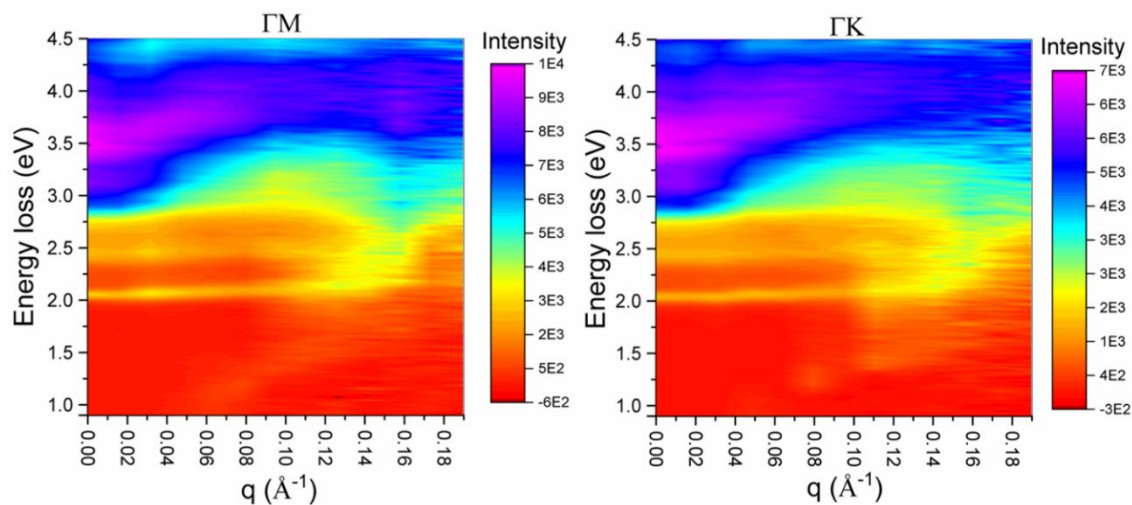


Figure S1. The q -E diagrams of monolayer WS₂ along Γ M and Γ K directions. No obvious in-plane anisotropy was found for all excitons.

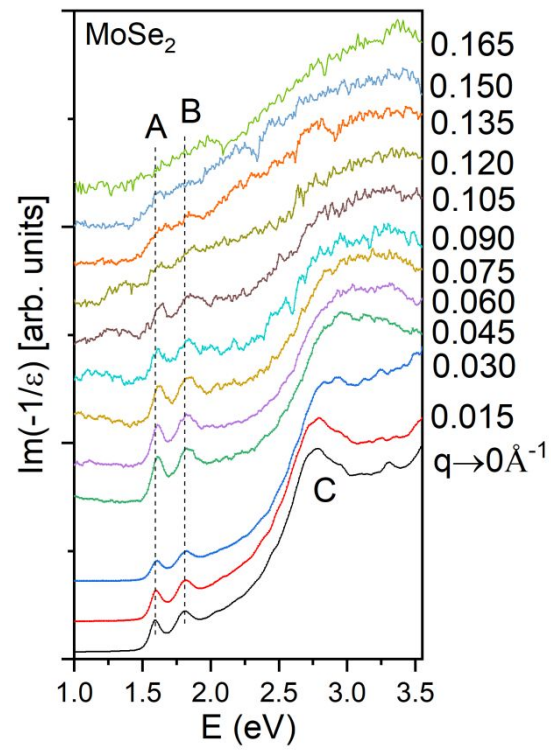


Figure S2. Experimental q -EELS loss functions of monolayer MoSe₂ along Γ K direction.

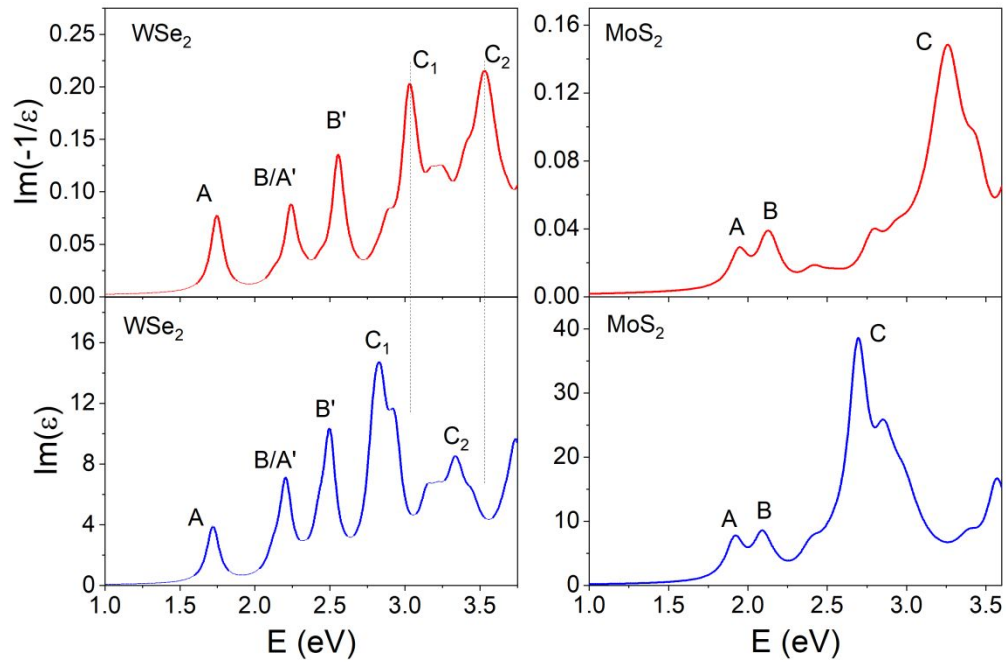


Figure S3. Theoretical EELS loss function $\text{Im}(-1/\epsilon)$ and optical absorption $\text{Im}(\epsilon)$ of TMDCs calculated by BSE+SOC at zero- q excitation. Note that the peaks in the optical absorption always have lower energy than their EELS counterparts, which is common according to the Kramers Heisenberg relation.

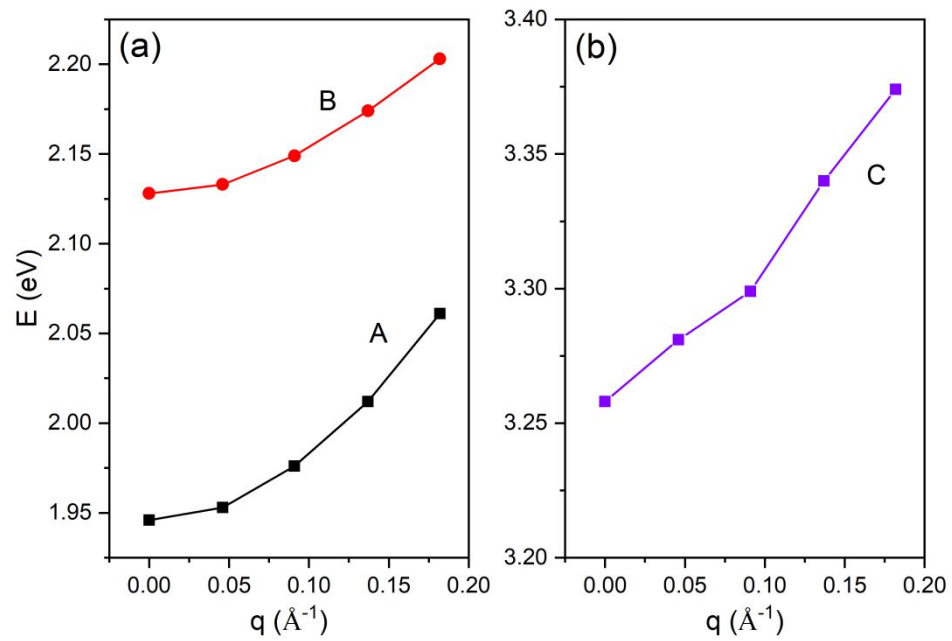


Figure S4. Theoretical exciton dispersion of MoS₂ derived by q dependent BSE loss functions.

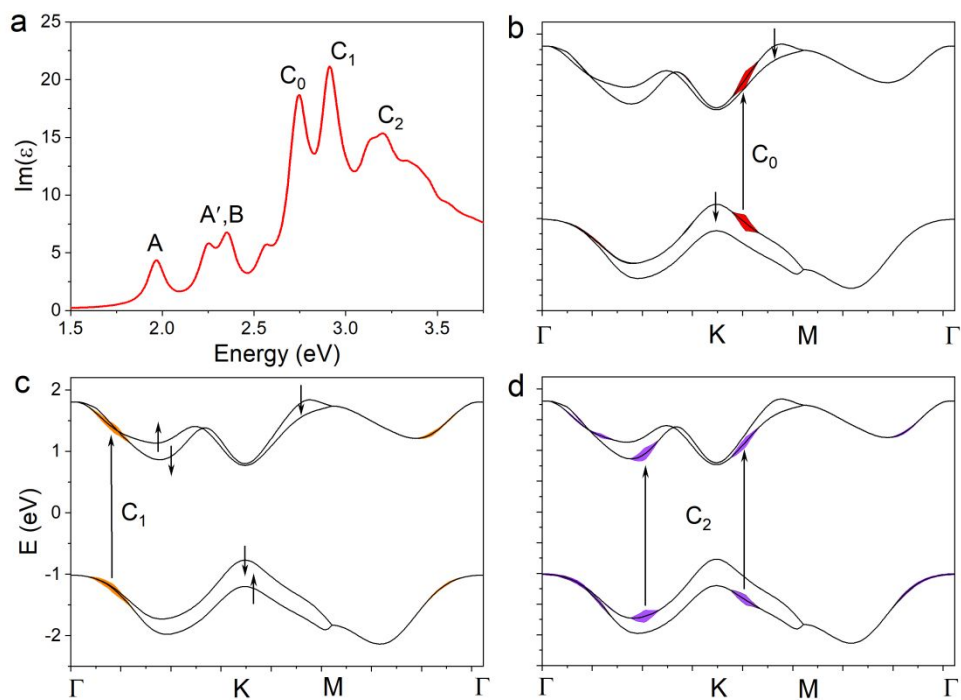


Figure S5. (a) BSE calculated absorption of WS₂ monolayer. (b-d) Excitonic weights projected onto the band structure of WS₂ with SOC included. The short arrows indicate the spin polarizations of each band, and long arrows show the transitions responsible for the multi-C peaks.

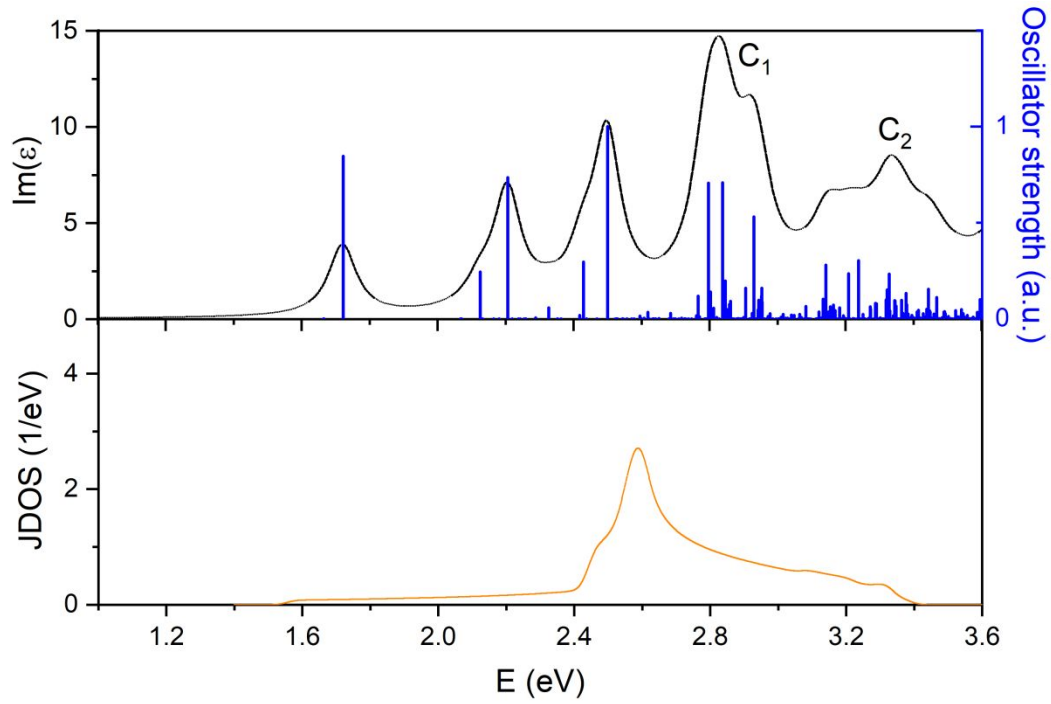


Figure S6. Oscillator strength (blue vertical bars) of each exciton in monolayer WSe₂. Note the absorption $\text{Im}(\epsilon)$ show intense peaks with large oscillator strength situated at energy positions far away from that of the DFT calculated JDOS peak. This is different from the case in Mo-DCs, where the band-nesting C peak results from the amplification of the high JDOS (joint density of states).

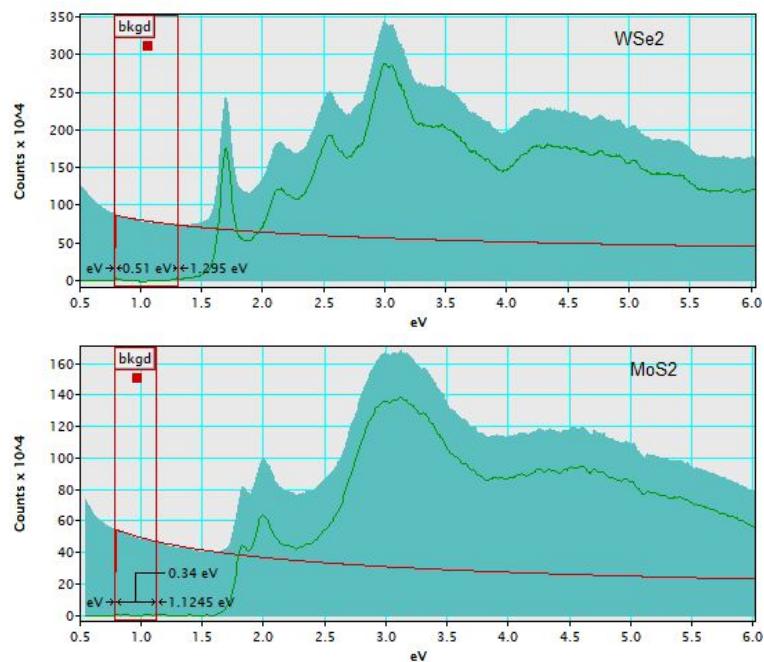


Figure S7. Power-law fitted removal of the zero-loss peak (ZLP) tail. Actually, the peak positions and line shapes will not be affected whether one remove the ZLP-tail background or not.