Revealing the Energy and Spatial Distribution of Edge States and Exciton Complexes in Transition Metal Dichalcogenides Using Multiphoton Microscopy

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Atomically layered two-dimensional (2D) transition metal dichalcogenides (TMDs) possess fascinating properties like strong many-body Coulomb-mediated interactions. The formation of multi-particle (e.g., two-particle excitons, three-particle trions and four-particle biexcitons) is the manifestation of the interactions [1,2]. But the phenomenon is generally observed under ultra-low temperature and is time-consuming for mapping the whole spatial distribution of exciton complexes by photo-luminescence (PL) spectrometer [1-4]. Multiphoton microscopy provides fast and straight-forward optical methods for determining the crystal orientation by large-area mapping and is a highly efficient method for detecting exactly structural features of TMDs [3]. In this work, we investigate the second-harmonic generation (SHG) of exciton-complex energies in WS_2 and WSe_2 monolayers with varying excitation wavelength and the SHG enhancement changed with different location. More significantly, we have found that the results of SHG are reliable by the comparison with PL mapping for exciton and trion, but not for biexciton. Furthermore, greatly enhanced SHG of one-dimensional atomic edges of MoS_2 monolayers is also observed owing to the characteristic called edge states caused by varied electronic structures at the different edge structure [5]. However, this phenomenon is not observed in WS_2 and WSe_2 monolayers indicating critical film quality requirements. This precise and practical multiphoton technique establishes a convenient route to characterize and optimize the potential for TMDs.

References

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[2] M. Barbone et al., Nat. Commun. 9 (2018) 3721
[3] L. Karvonen et al., Nat. Commun. 8 (2017) 15714
[4] H. Y. Jeong et al., Adv. Mater. 29 (2017) 1605043
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Figures

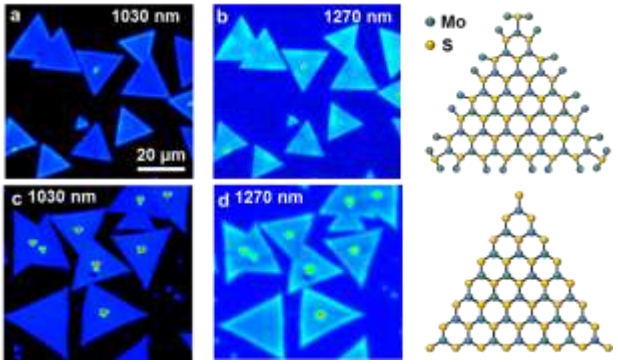

Figure 1: SHG images of (a, b) S-Mo Klein and (c, d) S-zz edge MoS_2 monolayers pumped at 1030 and 1270 nm, respectively.

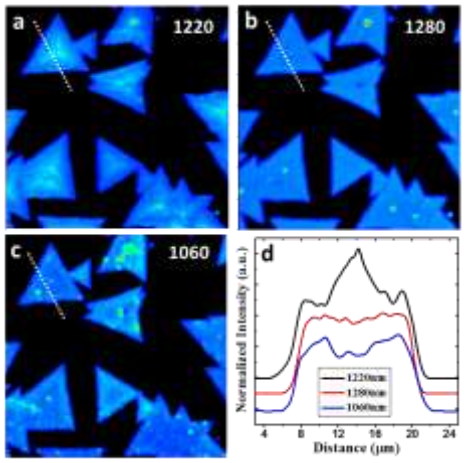

Figure 2: SHG images of WS_2 monolayers pumped at 1220 (a), 1280 (b) and 1060 nm (c), respectively. (d) Intensity profiles measured along the white dotted lines.