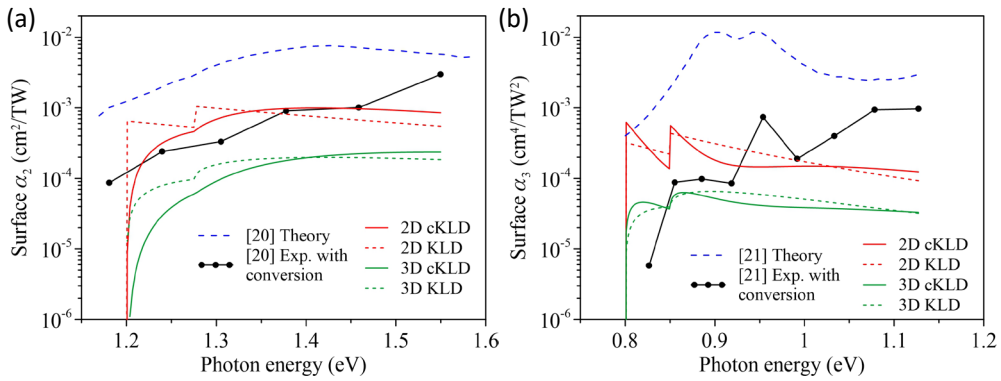

Figure 1: Internal surface 2PA absorption coefficient α_2 (a) and surface 3PA absorption coefficient α_3 (b) for monolayer MoS_2. Black dots and blue dashed curves are respectively experimental data and their theoretical prediction from [20, 21], both of which are converted to internal surface values. The red solid (dashed) curves are our theoretical predictions based on the 2D cKLD (KLD) formula. The predictions of 3D cKLD (KLD) after scaling by a material thickness of 0.63 nm are plotted as the solid (dashed) green curves for comparison. All KLD and cKLD curves employ two valence bands and a 2× degeneracy for K and -K valleys.

22. F. Zhou, J. H. Kua, S. Lu et al., "Two-photon absorption arises from two-dimensional excitons," Opt. Express **26**, 16093-16101 (2018).
23. P. Gong, H. Yu, Y. Wang et al., "Nonlinear optics in the electron-hole continuum in 2D semiconductors: two-photon transition, second harmonic generation and valley current injection," Science Bulletin **64**, 1036-1043 (2019).

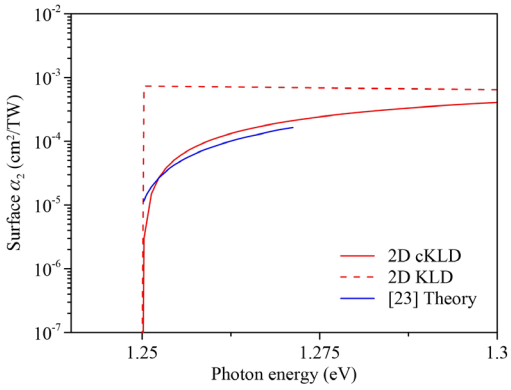

Figure 2: The internal surface 2PA absorption coefficient α_2 for WS_2 monolayer with a bandgap of 2.5 eV, calculated using 2D perturbation theory without electron-hole interaction (blue) [23], and our 2D cKLD (red solid) and KLD (red dashed) formulas.

23. P. Gong, H. Yu, Y. Wang et al., "Nonlinear optics in the electron-hole continuum in 2D semiconductors: two-photon transition, second harmonic generation and valley current injection," Science Bulletin **64**, 1036-1043 (2019).

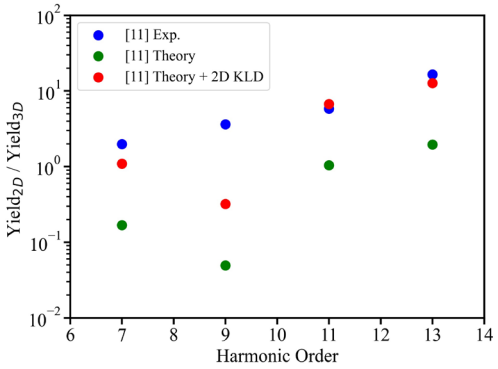

Figure 3: The ratio of HHG yields in monolayer MoS_2 and in a single layer of bulk MoS_2 as a function of the odd harmonic order. The blue dots are from the experiment of [11], the green dots are from the theory of [11], which considers only the intraband motion of conduction electrons[11], and the red dots are from the combination of the theory of [11] and the tunneling electron density ratio predicted by our 2D KLD theory.

11. H. Liu, Y. Li, Y. S. You et al., "High-harmonic generation from an atomically thin semiconductor," Nature Physics **13**, 262-265 (2017).