Atypical exciton-phonon interactions in WS2 andWSe2 monolayers: an ab-initio study

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Abstract The resonant Raman spectra of single-layered WS_2 and WSe_2 have been measured in a wide range of energies (using more than 25 laser lines). The resulting Raman excitation profiles of these very similar materials in both crystal and electronic structure show unexpected differences. All Raman features of WS_2 monolayers are enhanced by the first-optical excitations, but the response is not symmetric for the spin-orbit related X_A and X_B excitons. More interestingly, first order Raman bands of WSe_2 are not enhanced at $X_{A/B}$ energies, but they are at the X_C excitation. In this work, such intriguing phenomena are investigated by DFT calculations including excitonic effects by solving Bethe-Salpeter equation. We show that the ratio of the interaction of the X_C to the X_A excitons with the different phonons explains the different Raman responses of WS_2 and WSe_2 and the relative low Raman enhancement for the WSe_2 modes at $X_{A/B}$ energies (see the figure). These results reveal unusual exciton-phonon interactions and open new avenues for understanding the physics of 2D materials, where weak screening plays a key role coupling different degrees of freedom (spin, optic, electronic).

References

[1] E. del Corro, et. al. (submitted)
[2] Yilei Li, et al., Phys. Rev. B. **90** (2014) 205422

Figures

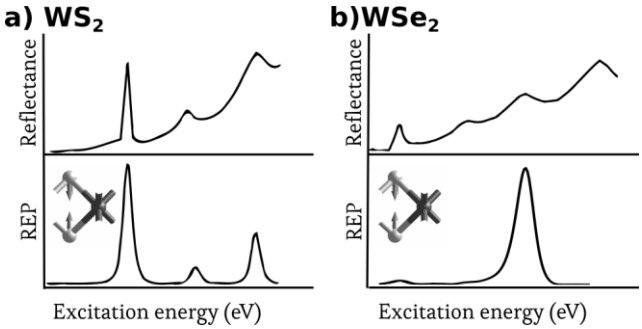

Figure 1: Comparison between the Raman excitation profile (REP) and the reflectance (adapted from [2]) for WS_2 and WSe_2, showing qualitative differences.