Temperature dependence of the upconversion photoluminescence in monolayer WS_2

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Photon upconversion is an anti-Stokes process in which an absorption of a photon leads to a reemission of a photon at energy higher than the excitation energy. The atomically thin semiconductors based on transition metal dichalcogenides are particularly promising for room temperature upconversion due to their very strong photon – exciton and phonon – exciton interactions.

Here, we demonstrate room temperature upconversion photoluminescence process in a monolayer semiconductor WS_2, with energy gain up to 150 meV. We attribute this process to transitions involving trions (T) and many phonons and free exciton complexes (X) [1]. We also show that the energy gain significantly depends on the temperature and increases from 42 meV at 7 K to 150 meV at 295 K. In order to gain insight into the temperature dependence of the mechanism of the upconversion emission, we compare the normal (PL) and upconverted photoluminescence (UPC PL) of the $WS_2/SiO_2/Si$ and $WS_2/hBN/SiO_2/Si$ structures at low, intermediate and high temperatures. At 7 K (Fig. 1 a, b) the energy gain of UPC emission amounts about 42 meV, which is comparable with the energy difference between the X and T emission lines and also nearly resonates with the energy of one optical phonon (A'_1 or E'). This suggests that at low temperature the UPC process is related to the coupling between the T and X states mediated by one optical phonon. At 70 K (Fig. 2 a, b), the exciting photon energy required to achieve a detectable upconversion photoluminescence is found to exceed E_{ex} = 2.009 eV. The higher energy gain of ~60 meV at 70 K suggests that more than one phonon is involved in the upconversion process.

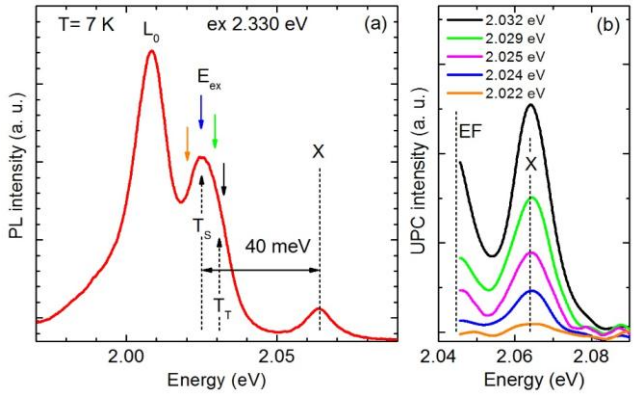

Fig. 1 (a) The PL spectrum recorded at 7 K. Colored arrows point out excitation energies used in UPC experiment. (b) The UPC spectra.

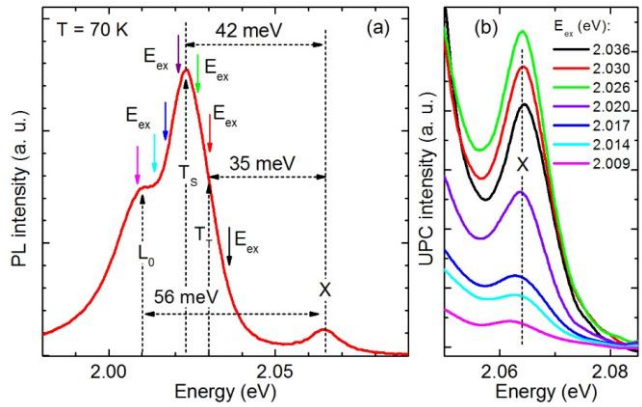

Fig. 2 (a) The PL spectrum recorded at 70 K. Colored arrows point out excitation energies used in UPC experiment. (b) The UPC spectra.

[1] J. Jadczak et al., *Nature Communications* **10**, 107 (2019).