Efficient high temperature multi-phonon assisted up-conversion photoemission in WS_2 monolayer

J. Jadczak [1], J. Kutrowska-Girzycka [1], P. Hawrylak [2] and L. Bryja [1]

[1] *Department of Experimental Physics, Wrocław University of Science and Technology, Wrocław, Poland*

[2] *Department of Physics, University of Ottawa, Ottawa K1N 6N5, Ontario, Canada*

The up-conversion (UPC) photoemission is an underlying process of light–matter interaction in which the emitted photon energy is higher than its excitation counterparts. This physical phenomenon is of fundamental importance for optical cooling in solids, in which UPC is related to anti-Stokes process enabled either by rare earth atoms or electron-phonon coupling [1]. Here, we demonstrate an efficient UPC photoemission process in monolayer WS_2 with an energy gain up to 155 meV at T = 295 K. We attributed this process to the multi-phonon resonant Raman scattering, where the absorbed and emitted photons are in resonance with the strongly localized and free excitons. These achievements is very promising for energy harvesting and application in optoelectronics.

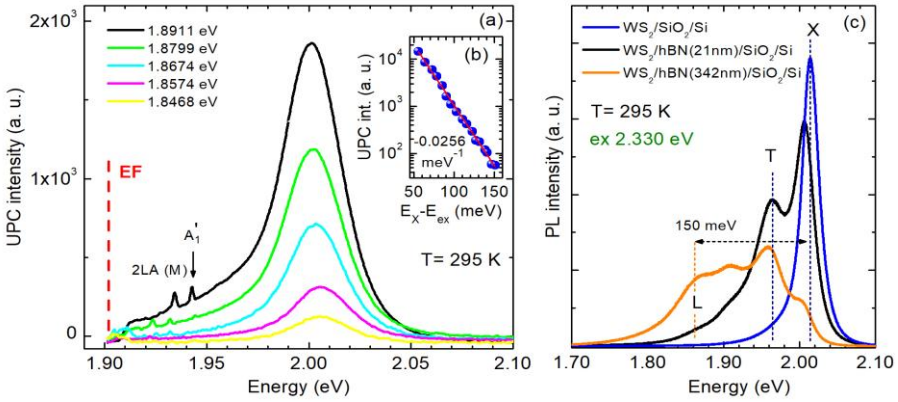

Fig. 1 (a) Examples of the UPC PL spectra. b) The dependence of the UPC integrated intensity on the (E_X – Eex). c) PL spectra recorded for WS_2 deposited on SiO_2/Si and on hBN/SiO_2/Si.

The highest efficiency UPC was observed and investigated for WS_2 MLs exfoliated on SiO_2/Si substrate. Fig. 1a presents the variations of the UPC emission spectra as a function of an incident photon energy. For all excitation photon energies one line, related to the neutral exciton (X) is observed at ~2.008 eV. The UPC intensity strongly depends on the excitation photon energy, whereas spectral shape and peak position remain nearly intact upon excitation energy variations. For the energy difference of the exciton and the excitation photon (E_X-E_{ex}) from 50 to 155 meV the integrated UPC PL intensity decreases with decreasing excitation photon energy with the rate equals -0.0256 meV^{-1} (Fig 1 (b)). The threshold excitation photon energy required to achieve observable UPC amounts to ~1.85 eV, which corresponds to PL energy of localized excitons L, which are distinguishable in the normal PL spectra even at *T*=295 K for the WS_2 monolayers exfoliated on hBN layers. Fig. 1c shows PL spectra from WS_2/hBN/SiO_2/Si with different thicknesses of hBN layers. For all structures the X line redshifts about 8 meV below to the its position recorded for the WS_2 ML deposited on SiO_2/Si. Moreover, an additional lines appear in the PL spectra. The transition at energy 1.964 eV, 42 meV below that of the X, is attributed to a negative trion (T), whereas the lower-energy, broad features L at ~1.86 meV are attributed strongly localized excitons.

[1] A. M. Jones at al. Excitonic luminescence up-conversion in a two-dimensional semiconductor. *Nature Physics* 12, 323–327 (2016).