Identification of low-temperature photoluminescence peaks by laser treatment in van der Waals epitaxially grown WS2 monolayers

ABSTRACT n° 11377

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Type of presentation : : Oral Communication

Domain : 2D materials and Van der Waals heterostructures

Date of submission : 2018-03-17

[397 Words]

Text

Transition metal dichalcogenides (TMDs) monolayers (MLs) are two-dimensional semiconductors that have been intensively studied over the past years due to spin-valley coupled physics and excitonic effects. Despite the considerable interest in TMDs, the detailed nature of the emission bands, especially of WS2 and WSe2 monolayers, still presents open questions. Besides, the synthesis and optical characterization of large-scale homogeneous monolayer samples represent an important step towards applications employing these material systems.

In this study, we investigate the optical properties of large-scale WS2 monolayers with sizes up to 35x30 mm grown by van der Waals epitaxy (VdWE) on SiO2/Si substrates. We employ macro- and micro-photoluminescence (PL) measurements to study spatial homogeneity, temporal stability, and laser induced doping on the uncapped samples [1]. We show that, in addition to the large structural homogeneity, the optical emission properties are very sensitive to laser irradiation at low temperatures as expected [1]. We monitor the variation in time of the emission bands associated with exciton (X), trion (T1 and T2), bound excitons (XB), and localized states (LS). We observe that the PL spectrum as a whole evolves significantly in time, displaying the appearance of additional emission bands and the disappearance of others. We demonstrate that this time evolution is mainly due to laser induced doping which can occur via carrier injection from the substrate or surface desorption. In particular, we use this optical instability effect to attribute the so-labelled T2 emission to a second trion state instead of a biexciton state. We also show that the time scale of the instability depends on the laser power density and probing spot. For low excitation intensities and large probing spots, the timescale for changes is in the minutes range. For higher laser excitation intensities and smaller spots, the PL time evolution occurs in a few seconds and a clear redshift is observed. Under these conditions, at low temperatures, the spectra do not recover their original shape when the excitation intensity is reduced to the low excitation regime. We also detect blinking of sharp bound exciton emissions which are usually attractive for single photon sources. The time instability of large scale WS2 ML emission and its dependence on the probing spot size are particularly important for future devices of reduced dimensions, such as single photon sources based on TMDs.

[1] V.Orsi Gordo et al, Nanoscale 10, 4807 (2018).