Strong Photoluminescence Fluctuations In Laser-thinned Few-layer WS_2

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Transition metal dichalcogenides (TMDs) have recently become a worldwide subject of intensive optical studies. One of the astonishing properties of these materials is that their bandstructures undergo a transformation from indirect-bandgap to direct-bandgap when decreasing the number of layers in the crystal lattice, which is accompanied by a substantial increase of the photoluminescence intensity. Since up to now there are no well-established procedures of synthesizing large-area TMD monolayers, a very desirable would be an efficient and reliable method of fabricating them from bulk TMD flakes.

In this communication we present our results of μ-Raman and μ-photoluminescence (μ-PL) study of few-layer WS_2 flakes that have been locally thinned down by a focused laser beam. The flakes were obtained by means of standard exfoliation of a bulk crystal and then deposited on a Si/SiO_2 substrate. Their actual thickness was determined using optical microscopy. After initial characterization a certain number of WS_2 layers were locally removed with the aid of high-power laser light. In order to get full control over this process spatially-resolved Raman maps were recorded on the locations subjected to laser-thinning (a typical result obtained on a 3-layer flake is shown in Fig. 1). The observed redshift of about 1 cm^{-1}, present in the middle of the map, suggests that the investigated flake was locally thinned down by one layer. We found the Raman spectra in the middle of the laser-induced hole to be very similar to that of an unperturbed two-layer WS_2 film.

In order to verify the quality of the obtained structure low-temperature μ-PL experiments were performed.

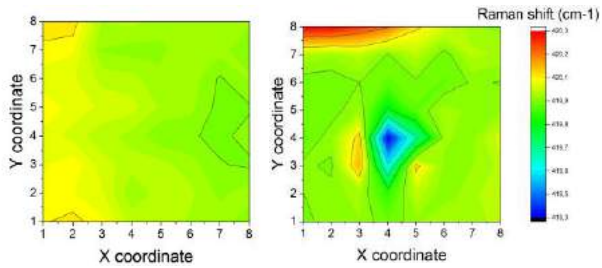

Figure 1. Spatially-resolved maps of the A_{1g} Raman mode of a 3-layer WS_2 flake before (left panel) and after laser-thinning (right panel).

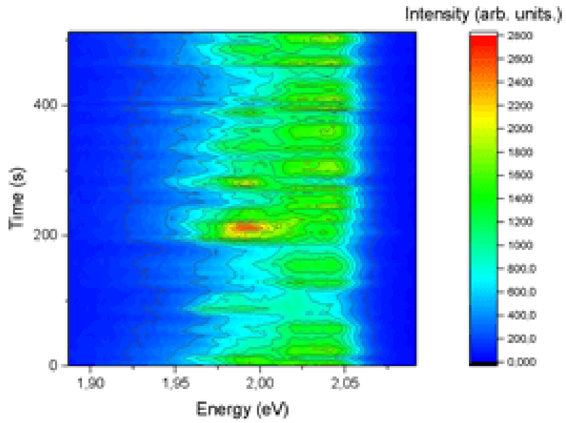

Figure 2. Subsequent photoluminescence spectra measured with 1 s acquisition time for laser excitation spot positioned at the boundary between the 3- and 2-layer parts of the investigated WS_2 flake.

It was found that the luminescence spectra measured outside the laser-thinned region were quite stable. Interestingly, huge intensity and energy fluctuations were detected at the boundary between the 3-layer area of the flake and the laser-thinned region (Fig. 2). Similar effects were found at the edges of a WS_2 monolayer flake, which has not been subjected to laser-thinning. The origin of the observed time evolution of the PL response will be discussed in terms of electrostatic potential fluctuations resulting from light-induced changes of the charge states of defects present in the laser-thinned area.