Lattice dynamics and photoluminescence emission of the two-dimensional $Mo_{1-x}W_xS_2$ alloys

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The family of the transition metal dichalcogenides (TMDs) such as MoS_2 and WS_2 have recently attracted a big attention due to two dimensional character of covalently bonded layers held together by weaker van der Waals forces. A single layer of MX_2 (M = Mo or W and X=S, Se or Te) consist of one atomic layer of metal atoms hexagonally packed between two trigonal atomic layers of chalcogenide atoms. In single-layer form the lack of inversion symmetry leads to remarkable optical and electronic properties different from those of bulk form. Unlike different 2D crystals, such as graphene and boron nitride, they are semiconductors, hence they reveal properties more attractive for specific application e.g. in optoelectronic devices.

In this work we focus on optical properties of $Mo_{1-x}W_xS_2$ monolayers. The studied flakes were mechanically exfoliated from bulk crystals grown by chemical vapour transport method (CVT) and transferred on Si/SiO_2 substrates. Prepared samples were identified and characterized by optical microscopy and atomic force microscopy (AFM). The micro-photoluminescence (μ-PL) measurements were performed for the wide range of temperatures (from 6 to 300 K). The power-dependent micro-Raman scattering investigations were carried out in backscattering geometry at room temperature under different conditions: ambient, acetone, water and saturated NaCl water solution. The excitation power varied from 12.5 μW to 1.25 mW and from 25 μW to 25 mW for the laser lines λ=633 nm and λ=532 nm, respectively.

The Raman spectra of the binary MoS_2 and WS_2 reveal two prominent first-order phonon modes: the E^1_{2g} and the A_{1g} modes. The E^1_{2g} mode is an in-plane vibration, for which the atoms are oscillating parallel to the basal plane of the van-der Waals coupled crystal layers. The A_{1g} mode is an out-of plane vibration, where the sulfur atoms are moving in opposite directions. In case of ternary materials the A_{1g} mode exhibits one-mode behavior whereas the E^1_{2g} mode exhibits two-mode behavior. We observe that in case of the WS_2 monolayer, similarly to MoS_2, the A'_1 mode shifts towards lower energies when the power density of the laser increases, whereas E' phonon mode remains essentially inert. This is likely due to stronger coupling of A'_1 mode with the excited d_{z^2} states. These results are consistent with previous studies of MoS_2 in FET geometry [1]. The comparison of the studied materials reveals that WS_2 is more sensitive to change of the carrier concentration than MoS_2. This tendency decreases linearly with increasing Mo content in $Mo_{1-x}W_xS_2$ alloys. Our results show that the charge carrier density can be effectively tuned by the light intensity.

[1] B. Miller, E. Parzinger, A. Vernickel, A. W. Holleitner, U. Wurstbauer, Appl. Phys. Lett. **106**, 122103 (2015).