



Carrier Density tuning in WS₂ monolayers via photochlorination

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Chlorine-doped tungsten disulfide monolayer (1L-WS₂) with tunable charge carrier concentration has been realized by pulsed laser irradiation of the atomically thin lattice in a precursor gas atmosphere. We have demonstrated the spatially resolved and reversible tuning of the PL properties of 1L-WS₂ using a pulsed laser photodoping process. The PL intensity can be significantly enhanced via the photo-assisted adsorption of p-type chlorine dopants to the monolayer lattice. This is due to the switching of the dominant PL process from the recombination of negative trions (X⁻) to that of neutral excitons (X), under the annihilation of residual electrons from the pristine crystal. DFT calculations confirm that chlorine adsorption can significantly affect the lattice electron density resulting in considerable shifts in the Fermi level position and to complete charge neutralization. Another remarkable finding is that the doping effect can be gradually reversed upon CW laser removal of the adsorbed chlorine species. This all-optical, bidirectional tuning of the electron density developed here can be advantageously used for spatially resolved doping modulation in 1L-WS₂ with micrometric resolution. It can therefore be important for the fabrication of TMD-based light-emitting and photovoltaic devices as well as in reducing the contact resistance between the TMD channel and the metal electrodes in field effect transistors. Although in the present study we have introduced a photochemical chlorination method to control the carrier density in 1L-WS₂, this process can be readily extended to alternate combinations of dopants and TMD materials.

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