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Excitons in two-dimensional transition metal dichalcogenides

Monolayers of transition metal dichalcogenides (TMDs) with MoS2 and WSe2 being the prominent examples are actively studied nowadays. These materials demonstrate remarkable optical properties controlled by the robust excitons, Coulomb-correlated electron-hole pairs, with the binding energies of several hundreds millielectronvolts. In my talk, the manifestations of excitons in linear and nonlinear optical properties of TMD monolayers are discussed. We start with the analysis of excitonic series in atomically thin semiconductors. Usually, the Wannier-Mott excitons form a series of 1s, 2s, 2p, … hydrogen-like states. In TMD monolayers the binding energies of the excitonic states strongly differ from the two-dimensional hydrogen atom model. This is mainly due to the specifics of the dielectric screening of the Coulomb interaction. Next, we briefly address the exchange interaction between the electron and the hole in the exciton. It is significantly stronger than in conventional semiconductor quantum wells. It provides efficient mechanism of the spin and valley decoherence of excitons. We also discuss the fine structure of the optically bright and dark excitons resulting from the combined effect of the exchange in spin-orbit interactions. Low (D3h) point symmetry of the monolayer with only three-fold rotation axis and without an inversion center leads to the mixing of p- and s-shell excitons with odd and even envelope functions. The microscopic model of the mixing is presented. This mixing impacts both linear and nonlinear optical properties. Particularly, it leads to the second harmonic generation in TMD monolayers. The nonlinear susceptibility responsible for the second harmonic generation is calculated. In agreement with experiment we observe giant enhancement of the second harmonic generation efficiency at the excitonic states. Finally, we briefly discuss exciton photoluminescence upconversion in TMD monolayers observed under excitation of 1s exciton state. Surprisingly the experiments on WSe2 monolayer reveal emission at higher energies corresponding to the excited 2s exciton as well as to the B:1s exciton associated with the spin-split valence subband. The effect results from the two-photon absorption via real intermediate states assisted by the Auger-like exciton-exciton scattering process. We also discuss possible manifestations of the bosonic effects in upconversion processes.