Interactions and Ultrafast Dynamics of Exciton Complexes in a Monolayer Semiconductor with Electron Gas

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Understanding the fundamental properties of exciton physics in the presence of 2D carrier gas in Transition Metal Dichalcogenides (TMDs) has been the subject of interest for many works in the recent years. The development of electrostatically-gated heterostructures of these materials facilitated precise control of free carrier density, which opened the way for the fundamental studies of strong exciton-carrier interactions present in TMDs, while also leading to such milestone achievements in the field of correlated electron systems like the observation of the Wigner crystal phase or the significant enhancement of the magnetic susceptibility. Despite, however, all of this progress, the particular influence of the Fermi sea of carriers on the optical response and dynamics of the exciton states, especially in the ultrafast limit, remains largely unexplored, rarely venturing beyond theoretical considerations.

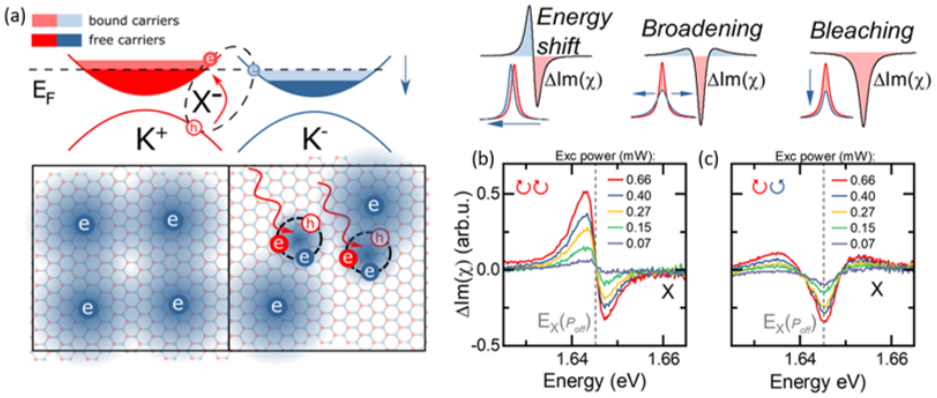

Fig. 1. : (a) Schematic of the influence of charged exciton photocreation on the electron gas and the related optical response of the neutral exciton in the same (b) and opposite (c) K-valley

We present femtosecond pump-probe measurements of neutral and charged exciton optical response in monolayer $MoSe_2$ to resonant photoexcitation of a given exciton state in the presence of 2D electron gas[1]. We show that creation of charged exciton (X^-) population in a given K^+, K^- valley requires the capture of available free carriers in the opposite valley and reduces the interaction of neutral exciton (X) with the electron Fermi sea. We also observe spectral broadening of the X transition line with the increasing X^- population caused by efficient scattering and excitation induced dephasing. From the valley-resolved analysis of the observed effects we are able to extract the spin-valley relaxation times of free carriers as a function of carrier density. Moreover, we analyze the oscillator strength and energy shift of X in the regime of interaction with electron Fermi sea under resonant excitation. From this we can observe the process of X decay by radiative recombination paired with trion formation. We demonstrate an increase of neutral exciton relaxation rate with the introduction of Fermi sea of electrons. We ascribe the observed effect to the increased efficiency of the trion formation, as well as the radiative decay caused by the screening of disorder by the free carriers.

References

[1] A. Rodek et al., Nanophotonics **13**, 487-497 (2024).