Abstract

Lead halide perovskite (LHP) has stimulated a plethora of research in photovoltaic community by attaining high 25.7% solar conversion efficiency due to extraordinary optical and electronic properties with low cost and easier processability. Two-dimensional (2D) LHP exhibits strong light-matter coupling and possesses even higher tunability and excellent photo-physical properties compared to their bulk counterparts. Still, question remains, what is the true timescale of carrier cooling and the debated mechanism underlying carrier relaxation. Also, a deep understanding and control of exciton interaction in 2D materials are of immense importance to unlock their use in technological applications.

The present thesis primarily deals with ultrafast carrier/quasiparticle dynamics in two different classes of perovskites: bulk organic-inorganic $CH_3NH_3PbI_{3-x}Cl_x$ mixed halide perovskite (MHP) and 2D all-inorganic $CsPbI_3$ nanosheets (NSs), using transient absorption spectroscopy. First, a complete ultrafast photo-physical insight of light matter interaction is investigated in MHP with the efficient recombination pathways. MHP shows the dominant free carrier nature and possesses a long carrier diffusion length of ~10 μm, high radiative efficiency, and two distinct direct band transitions which have not been observed earlier. Second, slow carrier relaxation mechanism in MHP is thoroughly investigated. The prolonged relaxation process over tens of picoseconds timescale is the effect of reduced energy loss rates due to quantitative interplay between hot phonon effect in early time, <5 ps, and Auger-heating effect on longer time, >10 ps. Finally, ultrafast photo-physical study of light-matter interaction is explored in NSs and a carrier dynamic model is proposed. Excitonic interaction dominates the optical response, unlike MHP, due to the presence of strong quantum confinement effect in NSs structure. The exciton−exciton interaction becomes stronger with higher hot-excitons concentration and thus, the excitonic coupling can be controlled by tuning the excitation photon density. Also, NSs show three distinct direct band electronic transitions with significantly different excitonic behaviors. This thesis provides profound aspects of ultrafast carrier and quasiparticle dynamics in bulk MHP and 2D NSs perovskite. Bulk MHP has proved to be a strong candidate for hot carrier solar cell due to delayed carrier relaxation, whereas, 2D perovskite is better suitable for advanced optoelectronic application.

Keywords: Lead halide perovskite, 2D nanosheets, Exciton/Carrier dynamics, Transient absorption spectroscopy, Hot carrier solar cell, Diffusion length, Auger-heating.