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Title:	Ultrafast Charge Transfer and Relaxation Dynamics in Metal Chalcogenide Based p-n Semiconductor Heterojunctions.
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Abstract:	<p>Metal chalcogenides are emerging solar energy materials and are blessed with unique properties such as high abundance and absorption coefficient (<math>10^4 - 10^5 \text{ cm}^{-1}</math>), tunable bandgap, low cost, and high stability, etc. Recently, metal chalcogenide-based p-n heterostructures have indeed gained tremendous attention in the field of solar energy conversion due to their potential for high efficiency. The p-n junctions are the fundamental and elementary building blocks of many electronic and energy storage devices, gas sensing devices, photovoltaics, photocatalysis viz. solar cells, diodes, light emitting diodes (LEDs), etc. They are the main active sites, where the efficient charge transfer processes take place. They offer fast separation and migration of the photoinduced charge carriers due to the presence of the built-in electric field. The efficient separation and longer lifetime of photoinduced charge carriers are crucial for any device. As the efficiency of any device is highly dependent on the dynamics of photoexcited charge carriers at the heterointerface, thus we must investigate excited state photophysical processes like charge transfer and recombination dynamics at the interface to fabricate efficient optical devices. Since these events are ultrafast in nature, we have utilized transient absorption spectroscopy (TAS) in my thesis work to track the interfacial charge transfer and relaxation events that occur over time scales of a few femtoseconds (fs) to nanoseconds (ns) in different heterojunction systems. To design cost effective devices, it is vital to comprehend the dynamics of these materials in their excited states. My first work deals with the synthesis of <math>\text{Cu}_2\text{ZnSnS}_4</math> (CZTS) nanoparticles and CdS quantum dots (QDs) using the hot-injection method followed by fabrication of CZTS, CdS, and CZTS/CdS heterojunction thin films with the help of a spin-coating technique. Steady state and time-resolved photoluminescence studies confirm the hole transfer from photoexcited CdS to CZTS and the hole transfer rate constant was calculated to be <math>0.366 \times 10^9 \text{ sec}^{-1}</math>. The ultrafast studies clearly suggest electron transfer from the CZTS domain to the CdS domain with a time constant of 0.14 ps, establishing the charge transfer efficiency in the heterointerface. Thus, our results demonstrate the fast charge separation in CZTS/CdS heterojunction film. The next work is designed to explore the underlying hot carrier relaxation and transfer strategies within the SnSe/CdSe p-n heterostructure. SnSe/CdSe heterostructure is synthesized by hot injection followed by the cation exchange method. The epitaxial growth of the (400) plane of SnSe along the (111) plane of CdSe has been confirmed through HRTEM analysis. Transient absorption spectroscopy (TA) studies demonstrate a drastic enhancement of the CdSe biexciton signal which points toward the hot carrier transfer from SnSe to CdSe in a short time scale. Afterward, these carriers are transferred back to SnSe. The observed delocalization of carriers in these two systems is crucial for an optoelectronic device. Further, we examine the ultrafast excited-state dynamics at the junction between Sb<sub>2</sub>Se<sub>3</sub> and CdSe QDs in the Sb<sub>2</sub>Se<sub>3</sub>/CdSe p-n heterostructure system. The enhanced intensity of the CdSe hot excitonic (1P) bleach in the heterostructure system confirmed the hot electron transfer, instead of getting trapped in the defect states present in Sb<sub>2</sub>Se<sub>3</sub>, migrate from Sb<sub>2</sub>Se<sub>3</sub> to the CdSe system. Further, both the 1S and 1P signals are dynamically very slow in the heterosystem, validating this charge migration phenomenon. Interestingly, recovery of the 1P signal is much slower than that of 1S which indicates the robustness of hot electron transfer in this unique heterojunction, that helps in increasing the carrier lifetime in the hot state. My next work is based on studying the interfacial charge transfer processes at the p-n junction of the 1D/0D SnS@CdS p-n heterostructure system. The structural and morphological properties presented that CdS QDs are uniformly anchored on the surface of SnS NRs resulting in an intimate contact between two components. The steady state and time resolved photoluminescence analysis demonstrated the transfer of photoexcited holes from CdS QDs to SnS NRs which was further confirmed by transient absorption studies. Moreover, the transient studies reveal the delocalization of electrons between the conduction band of SnS NRs and CdS QDs in the SnS@CdS heterostructure, resulting in efficient charge separation across the p-n heterojunction. Finally, we have explored the charge transfer and recombination dynamics in Sb<sub>2</sub>Se<sub>3</sub>/CdS thin film p-n heterojunction. The Sb<sub>2</sub>Se<sub>3</sub>/CdS heterojunction film was fabricated using the thermal evaporation technique. The ultrafast studies reveal the electron transfer from Sb<sub>2</sub>Se<sub>3</sub> to CdS thin film and spatial charge separation at the Sb<sub>2</sub>Se<sub>3</sub>/CdS p-n junction. Moreover, the photocurrent response of Sb<sub>2</sub>Se<sub>3</sub>/CdS heterojunction was greatly enhanced as compared to bare Sb<sub>2</sub>Se<sub>3</sub> and CdS films under light illumination owing to improved separation rate of photogenerated charge carriers in heterojunction system, which correlates with our transient investigations, and are crucial factors for the fabrication of solar energy-based devices. In conclusion, we have investigated the intrinsic photophysical processes at the interface of some very prominent metal chalcogenides, which will open a new route for the fabrication of extremely effective photovoltaic and photocatalytic devices.</p>
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