Influence of Chemical Composition on the Relaxation Rates of Charge Carriers in PbX/CdX, X = S or Se, Core/Shell Quantum Dots

Svetlana Kilina, Levi Lystrom, Patrick Tamukong

North Dakota State University
Chemistry and Biochemistry Department

Colloidal quantum dots (QDs) have been heavily investigated for a range of applications including display technology, bio-imaging, and solar energy conversion. Traditional QDs are highly sensitive to environmental factors arising from dynamic passivation of surface atoms by ligands. Heterostructures nanoparticles such as core/shell QDs have been shown to reduce the sensitivity of the QD to environmental factors. Core/shell QDs are comprised of two semiconductors, in this work we study pseudo-type II (PbX)16/(CdX)52, X = S or Se, core/shell QDs. The chemical composition of the QDs control the excited state properties via manipulation of the ground state electronic properties. For solar energy conversion, the electronic properties of the pseudo-type II QDs can dramatically reduce the rate of charge carrier cooling. Non-adiabatic molecular dynamics simulations were performed showing that PbS/CdS charge carriers cool in approximately half the time compared to PbSe/CdSe QDs, which can be attributed to the reduced vibrational frequency of the metal chalcogenide bond vibration in PbSe/CdSe QDs. The reduced charge carrier cooling can aid in the efficiency of the solar cell by improving Carrier Multiplication (CM), generating multiple pairs of charge carriers from a single photon. CM must occur quicker compared to charge carrier cooling which makes PbSe/CdSe QDs are more suitable for solar energy conversion compared to PbS/CdS QDs.