

Excitation energy dependence of
efficiency of charge carrier
relaxation and photoluminescence
in colloidal InP quantum dots

Randy J. Ellingson, Jeff L. Blackburn,
Pingrong Yu, Garry Rumbles, Olga I.
Mičić, and Arthur J. Nozik

Center for Basic Science, National
Renewable Energy Laboratory, Golden,
Colorado 80401

Colloidal quantum dots offer great potential as candidates for charge transporters and light absorbers in photovoltaic solar cells. These materials are prepared in solution and are thus amenable to the armory of chemical synthesis methodologies to tune and improve their electronic properties as well as their processability. As these materials can be processed from solution, it opens up the opportunity to prepare photovoltaic solar cells that are both efficient and relatively cheap to manufacture.

This presentation will focus on the electronic properties of colloidal quantum dots made from the III-V semiconductor, indium phosphide, InP. The covalent nature of the bonding in these quantum dots makes them more difficult to synthesize than the more prevalent II-VI materials, such as cadmium selenide, CdSe. The band gap of these materials is dependent upon the size of the dots and therefore they can be tuned to optimize absorption of the solar spectrum. Alternatively, there is an opportunity to use the restricted carrier relaxation mechanisms in these quantum-confined systems to utilize photons absorbed at energies higher than the band gap. Two such examples are the extraction of the absorbed energy from the hot carriers prior to relaxation or the enhancement of intra-dot, impact ionization to increase the number of carriers per absorbed photon. To examine these processes in more detail requires knowledge of the excited state properties of the quantum dots and the relationship to their chemical make-up, particularly the nature of the surface. It is this topic that is focused on here.

Femtosecond transient absorption spectroscopy has been used together with time-integrated photoluminescence (PL) measurements to study charge carrier relaxation in colloidal InP quantum dots (QDs). Measurements of band-edge photoinduced bleaching as well as PL intensity measurements indicate that the fraction of charge carriers relaxing to the band edge states depends on excitation wavelength, with a markedly reduced relaxation efficiency observed for excitation well above the absorption edge. The results concur with previous research on CdSe and InP QDs, and suggest that with increasing excitation energy there arise relaxation pathways involving surface or external energy states exhibiting reduced radiative efficiency. Such highly-excited carriers are either inhibited or deviated from reaching the band edge states. Excitation intensity-dependent measurements indicate that those charge carriers that contribute to the band edge absorption bleaching also contribute proportionally to the time-integrated PL spectrum.