The Carrier Multiplication Controversy in Semiconductor Nanocrystals

Carrier multiplication (CM), the generation of multiple electron-hole pairs by absorption of a single photon, is a phenomenon that could lead to enhanced efficiency of solar-energy-conversion devices such as photovoltaics if materials could be engineered for high-efficiency CM. In bulk systems, the process occurs by impact ionization and is inefficient at visible photon energies. It has been argued that CM could be enhanced in semiconductor nanocrystal quantum dots (NQDs) due to potentially reduced rates of cooling of hot carriers by phonon emission resulting from large, confinement-induced intraband level splittings; relaxation of momentum conservation due to the finite spatial extent of NQDs; and reduced Coulomb screening. Indeed, CM efficiencies near the energy-conservation limit have been reported, but a controversy has arisen due to the tremendous inconsistency in the efficiencies reported by different groups and even the same group. I will describe recent experimental results that account for variations in reported CM efficiencies in terms of photoinduced charge separation, a process that was not previously accounted for in spectroscopic measurements of CM and that has implications beyond the study of CM. In light of these results, I will revisit the assumptions about the expected causes of enhanced CM in NQDs and assess the current state of the search for efficient CM in NQDs and other materials. Given that confinement increases the band gap of NQDs, I will also discuss how different materials should be compared in assessing whether CM is enhanced and their utility in devices intended to enhance solar energy conversion via CM.