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Seven Excitons per Single Photon Using Semiconductor Nanocrystals

VICTOR KLIMOV, Los Alamos National Laboratory

The efficient conversion of photon energy into electrical charges is a central goal of much research in physics, chemistry, and biology. A usual assumption is that absorption of a single photon by a material produces a single electron-hole pair (exciton), while the photon energy in excess of the energy gap is dissipated as heat. In 2004, we reported for the first time that nanocrystals (NCs) of PbSe could respond to absorption of a single photon by producing two or more excitons with the unity probability (Phys. Rev. Lett. 92, 186601, 2004). Our more recent findings indicate that this carrier multiplication process can generate multiple charges with quantum efficiencies that correspond to the ultimate limit dictated by energy conservation. For example, for photon energy of 7.8 energy gaps, a maximal possible number of photogenerated excitons based on energy considerations is 7, which is exactly the number measured in our experiments (Nano Lett. 6, 424, 2006). Another unexpected feature of carrier multiplication is that it results in unusual distributions of carrier populations that cannot be described by Poisson statistics. Specifically, by selecting certain photon energies, we obtain photoexcited NC ensembles with nearly pure single multiplicities (i.e., all excited NCs contain the same number of excitons) that can be tuned in the controlled way from 1 to 7 (Phys. Rev. Lett. 96, 097402, 2006). While the exact mechanism for carrier multiplication in NCs is still under debate, one factor, which likely contributes to high efficiencies of this process, is a unique property of the NCs to produce significant carrier-carrier interactions as indicated, e.g., by our previous Auger recombination studies (Science 287, 1011, 2000). This confinement-enhanced Coulomb coupling can lead to the unusual mechanism for direct photogeneration of multiexcitons via virtual single-exciton states, which can explain our observations of very short, sub-200 femtosecond buildup times of multiexciton populations in the regime of carrier multiplication (Nature Phys. 1, 189, 2005).