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Exciton Relaxation Dynamics and Ultra-Efficient Exciton Multiplication in Nanocrystals: A New Model and Relevance to Solar Photon Conversion

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In bulk semiconductors photoinitiated electron-hole pair multiplication can occur via impact ionization (I.I.). However, this process requires photon energies of 4-5 times the bandgap because of the need to conserve crystal momentum and the competing rate of phonon emission. This limits the application of I.I. to increase the solar conversion efficiency in photovoltaic devices. For nanocrystal QDs, these constraints are relaxed, and exciton multiplication is thus expected to be greatly enhanced (1). Greatly enhanced exciton multiplication in PbSe nanocrystals was first confirmed by Schaller and Klimov (2). We report ultra-efficient multiple exciton generation (MEG) in PbSe and PbS nanocrystals, and explain our results by a new theoretical model based on the coherent superposition of excitonic states whereby multiple excitons are created instantaneously upon photon absorption (3); such a coherent process has never been reported before in semiconductors. Astonishingly high quantum yields of 300 %, indicating creation of three excitons/photon for every nanocrystal, have been observed in PbSe at photon energies four times the energy gap. The new model predicts the occurrence of quantum beats between the coupled coherent states, and we observe such quantum beats in PbSe and PbS nanocrystals. Another prediction of the new model, supported by our data, is that the threshold photon energy for MEG in PbSe nanocrystals is twice its energy gap. These results have important implications for greatly improved optoelectronic devices. Refs: (1) A.J. Nozik, Physica E14, 115 (2002); (2) R. Schaller and V. Klimov, Phys. Rev. Letts. 92, 186601(2004); (3) R.J. Ellingson, M.C. Beard, P.Yu, O.I. Micic, A.J. Nozik, A. Shabaev, Al. L. Efros, submitted to Science (2004)

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