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Multiexciton absorption in CdSe nanocrystals[1] ALBERTO FRANCESCHETTI, YONG ZHANG, National Renewable Energy Laboratory — Efficient multiple-exciton generation (MEG) has been recently reported in semiconductor nanocrystals. In this process, a single absorbed photon generates two or more electron-hole pairs. The MEG efficiency has so far been evaluated assuming that the change (bleaching) of the absorption spectrum due to MEG is linearly proportional to the number of excitons (N_X) that are present in the nanocrystal. We have examined this assumption using atomistic pseudopotential calculations for colloidal CdSe nanocrystals ranging in size from 3 to 4.6 nm. We found that the bleaching of the first absorption peak, $\Delta\alpha_{1S}$, depends non-linearly on N_X, due to carrier-carrier interactions. When a single exciton is present in the nanocrystal, the 1S exciton peak is already 65-75% bleached. This non-linearity mandates an upper bound of 1.5 to the value of the normalized bleaching that can be attributed to MEG, significantly smaller than the limit of 2.0 predicted by the linear scaling assumption. Thus, measured values of the normalized bleaching in excess of 1.5 in CdSe nanocrystals cannot be due entirely to MEG, but must originate in part from other mechanisms.

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