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Photoinduced Electron Transfer to Engineered Surface Traps in CdSe Nanocrystals MARCO CALIFANO, University of Leeds, HAIMING ZHU, YE YANG, Emory University, KIM HYEON-DEUK, Kyoto University, NIANHUI SONG, Emory University, YOUWEI WANG, WENQING ZHANG, Chinese Academy of Sciences, OLEG PREZHDO, University of Rochester, TIANQUAN LIAN, Emory University — Quantum confined nanomaterials, such as semiconductor nanocrystals (NCs), have emerged in the past decade as a new class of materials for solar energy conversion. An appropriate model for describing photoinduced charge transfer in these systems is, however, still lacking. Recently we observed that the rate of photoinduced electron transfer from CdSe NCs to molecular acceptors increased with decreasing NC size (and increasing driving force) exhibiting a lack of Marcus inverted regime behaviour over an apparent driving force range of 0-1.3 V. Our atomistic semiempirical pseudopotential calculations show that an Auger assisted ET mechanism, in which the transfer of the electron is coupled to the excitation of the hole, can circumvent the unfavourable Frank-Condon overlap (that is a signature of inter- or intra- molecular electron transfer) in the Marcus inverted regime, reproducing our observed ET rates with remarkable accuracy. We conclude that electron transfer from quantum dots differs from electron transfer originating from both molecules and bulk semiconductors. It proceeds via a novel Auger-assisted pathway which we believe is available to most excitonic nanomaterials. This new finding will have a major impact on the design of next generation solar energy harvesting devices.

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