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Light Harvesting in Functionalized SiQD Assemblies via Spatially Separated Excitons HUASHAN LI, ZHIGANG WU, TIANLEI ZHOU, ALAN SELLINGER, MARK LUSK, Colorado School of Mines — Silicon quantum dots (SiQDs) with diameters less than 5 nm are particularly attractive for photovoltaic applications [1,2], but their optical gap is too large to match the solar spectrum. Although recent progress on solution processing techniques provides more opportunities for functionalizing SiQD, nontrivial absorption under 3 eV has yet to be achieved [3]. The absorption of photons through the direct generation of spatially separated excitons at dot-ligand interfaces may be a promising strategy for overcoming this challenge. We consider the idea computationally and show that it is indeed possible to capture photons of much lower energy using very small SiQD. The key is to establish a type-II energy level alignment in conjunction with strong electronic coupling between the dot and ligand. Our analysis indicates that conjugated vinyl bonds to common organic ligands satisfy both of these conditions. In principle, this allows the optical gap of SiQD to be tuned to arbitrarily small values independent of their size. For the prototype system of 2.6 nm SiQDs, we predict that triphenylamine (TPA) termination will result in a 0.47 eV redshift of the optical gap along with a boost of absorption intensity near the band edge, a result consistent with our experimental realization of the system. We will also discuss the results of a computational analysis of the robustness of the absorption spectrum against oxidation and extra alkyl ligands within this new paradigm. [1]Lin, Z. et al., ACS Nano 6, 4029, 2012. [2] Li, H. et al., ACS Nano 6, 9690, 2012. [3] Dung, M. X. et al., Chem. Asian J. 8, 653, 2013.

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