

Interfacial Charge Transfer from Cesium Lead Halide Perovskite Nanocrystals

David Nenon^{1,2} (presenting), Jacob Olshansky,^{1,2} A. Paul Alivisatos^{1,2}

¹Department of Chemistry, University of California, Berkeley, California 94720, United States

²Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, United States

Following the remarkable rise in performance of perovskite solar cells, the lead halide perovskite family has emerged as an important class of optoelectronic materials. More recently, all-inorganic nanocrystals (NCs) of cesium lead halide (CsPbX_3 , $X=\text{Cl}^-$, Br^- , I^-) perovskite have attracted attention for their high photoluminescence quantum yields, readily tunable halide composition, and versatile colloidal syntheses.

Unlike prototypical cadmium selenide NCs, perovskite NCs exhibit surprisingly high luminescence (PLQY >80%) without a passivating shell. [1] The absence of a shell means charges can be extracted without having to pass through a tunneling barrier, which is known to significantly reduce the charge transfer rate. [2] For these reasons, we have chosen to study hole transfer to surface-bound aminoferrocene ligands (Figure 1). By building upon a methodology developed in our group, we are able to measure the hole transfer rate for perovskite NCs of various sizes. Figure 1 shows that a relatively small change in NC size leads to a dramatic change in the hole transfer rate – the ability to significantly modulate the rate will be discussed further.

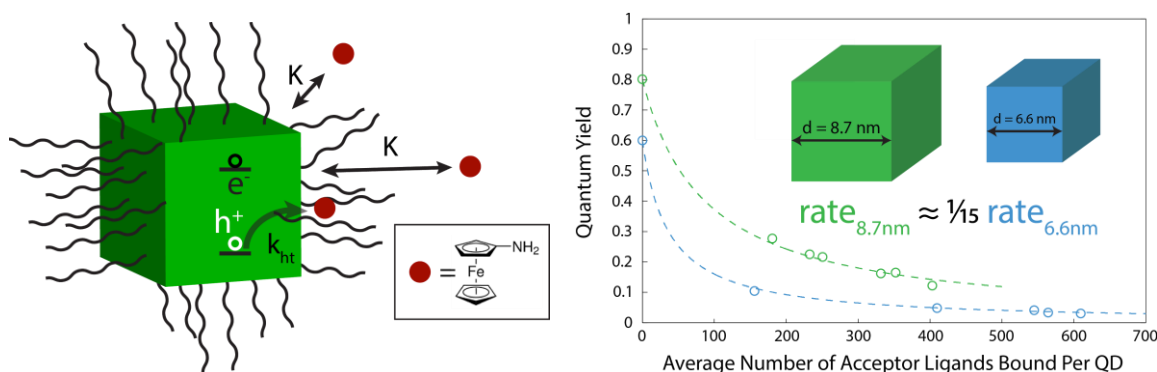


Fig. 1 Schematic of hole transfer from a CsPbBr_3 quantum dot to aminoferrocene ligands (left). Quantum yield as a function of bound acceptor ligands, N , for two CsPbBr_3 QD sizes (right).

Whereas the initial report of charge transfer from perovskite NCs to physisorbed acceptors determined the rates to be exceptionally fast, [3] we have found ourselves to be in a different regime. The rates we have measured to bound ligands are comparable to that of cadmium selenide with a cadmium sulfide shell thickness of a few monolayers, which is quite surprising given that the perovskite NCs are synthesized without a shell. The possible physical origins and implications of these modest rates will be discussed.

Finally, we have learned how to quantify the binding of non-native ligands on the dynamic surface of these nanocrystals, which has not yet been reported. This has important implications not only for accurately determining charge transfer rates, but for understanding the role of the surface in this novel system. Our investigations into the extraction of charges will inform the potential of these exciting new materials for energy conversion technologies and other applications.

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- 2) Ding, T. X.; Olshansky, J. H.; Leone, S. R.; Alivisatos, A. P. *J. Am. Chem. Soc.* **2015**, *137*, 2021-2029.
- 3) Wu, K.; Liang, G.; Shang, Q.; Ren, Y.; Kong, D.; Lian, T. *J. Am. Chem. Soc.* **2015**, *137*, 12792-12795.