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Designing and probing ultrafast energy/charge transfer kinetics in Ruddlesden-Popper perovskites and QDs attached photochromic devices

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The semiconductor perovskites materials and quantum dots (QDs) demonstrate outstanding optical properties. The perovskites L exhibit wide color emission that makes them the highly suitable candidate for LEDs and QDs exhibit narrow emission that makes them suitable for fluorescent probes. 2D organolead halide perovskites and their 3D analogous have exhibited encouraging performance metrics of LED, like low turn-on voltages and external quantum efficiencies. An optical cascading mechanism is observed in electrically emissive Ruddlesden-Popper (RP) perovskite series $((C_8H_{17}NH_3)_2(CH(NH_2)_2)_{m-1}Pb_mBr_{3m+1})$. A mixture of 3D formamidinium lead bromide CH(NH₂),PbBr₃ and 2D octyl ammonium lead bromide (C₈H₁₇NH₃)₂PbBr₄ perovskite (~0-80% 2D) are prepared to develop an understanding regarding the energy cascading mechanism. Ultrafast transient absorption and fluorescence techniques suggest energy transfer from high bandgap "donor" too low bandgap "acceptor" in RP perovskite films for 20% (2D) mixed RP perovskite. Following excitation at 400nm in RP very fast decay of 435nm exciton and very fast rise (390 ft) of 535nm bleaching signal suggest very efficient energy transfer from donor to acceptor for 20% 2D perovskite. By further increasing the concentration of 2D in 3D RP perovskites, energy transfer is delayed and it deteriorates the LED efficiency due to incomplete energy transfer. These findings shed light on the importance of engineering the acceptor to donor ratio to gain efficient energy transfer. Parallelly in II-VI semiconductor QDs attached photochromic azobenzene molecules a fast charge/energy transfer is observed. Upon UV irradiation, the photoresponsive azobenzene undergoes a reversible trans-cis isomerization. The photoinduced trans-cis transformation helps to transfer photoexcited charge/energy transfer from the conduction band of the QDs to the LUMO of the cis-isomer of the azobenzene. As a result, the fluorescence of QDs can be modulated to fabricate fluorescent probes for a wide range of optical applications.

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