Dimensional Deduction in Metal Halides: from Photovoltaic to Luminescent Materials

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Hybrid organic-inorganic lead halide perovskites have emerged as promising photovoltaic materials with the best solar cell efficiency over 25%. Dimensional reduction of 3D metal halides to 2D, 1D, and 0D further leads to the discovery of many highly efficient luminescent materials with photoluminescence quantum efficiency close to 100% [1]. In this talk, I will discuss electronic, dielectric, and defect properties that are responsible for the efficient carrier transport in hybrid halide perovskites [2] [3] as well as the microscopic origin of the efficient luminescence in low-dimensional metal halides [4] [5] based on hybrid density functional calculations. The ΔSCF method combined with the hybrid functional are used to probe the complex excited-state energy landscape of low-dimensional metal halides, leading to the discovery of multiple self-trapped excitons in several compounds [6][7]. Calculated emission energies of self-trapped excitons agree very well with experimental results and explain the optical emission trends observed in low-dimensional metal halides.

1. "Low Dimensional Metal Halide Perovskites and Hybrids", C. Zhou, H. Lin, Q. He, L. Xu, M. Worku, M. Chaaban, S. Lee, X. Shi, M. -H. Du, and B. Ma, Mater. Sci. Eng. -R: 137, 38-65 (2019).
2. "Efficient Carrier Transport in Halide Perovskites: Theoretical Perspectives", M. -H. Du, J. Mater. Chem. A, 2, 9091 (2014).
3. "Density functional calculations of native defects in $CH_3NH_3PbI_3$: Effects of spin-orbit coupling and self-interaction error", M. -H. Du, J. Phys. Chem. Lett. 6, 1461 (2015).
4. "Unraveling Luminescence Mechanisms in Zero-Dimensional Halide Perovskites", D. Han, H. Shi, W. Ming, C. Zhou, B. Ma, B. Saparov, Y. -Z. Ma, S. Chen, and M. -H. Du, J. Mater. Chem. C 6, 6398-6405 (2018).
5. "Impact of Metal ns^2 Lone Pair on Luminescence Quantum Efficiency in Low-Dimensional Halide Perovskites", H. Shi, D. Han, S. Chen, and M. -H. Du, Phys. Rev. Mater. 3, 034604 (2019).
6. "Microscopic Origin of Multiple Exciton Emission in Low-Dimensional Lead Halide Perovskites", M. -H. Du, J. Chem. Phys. 151, 181101 (2019).
7. "Emission Trend of Multiple Self-Trapped Excitons in Luminescent 1D Copper Halides", M. -H. Du, ACS Energy Lett. 5, 464-469 (2020).