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Photoluminescence in lead halide perovskites and the role of defects

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The simple processability of lead halide perovskite semiconductors, typically from solutions at temperatures close to room temperature, exposes this class of materials to a non-negligible level of unintentional structural and chemical defects. Ascertained that their primary optoelectronic properties meet the requirement for high efficiency optoelectronic technologies, a lack of knowledge of the nature of the defects and their role in the device operation currently represents a major challenge, limiting the further enhancement of the device efficiency and, importantly, their stability and reliability. Here, we investigate the role of defects in the photo-excitation dynamics in a variety of lead halide perovskite systems using novel optical and structural probes.

Firstly, we use excitation correlation photoluminescence (ECPL) spectroscopy [1] to investigate the recombination dynamics of the photo-generated carriers in 3D lead bromide perovskites and quantitatively describe the carrier trapping dynamics within a generalization of Shockley-Read-Hall formalism. The superior sensitivity of our spectroscopic tool to the non-linear carrier interactions enables us to identify the nature and energetics of the defects unambiguously. In the case of polycrystalline films, depending on the synthetic route, we demonstrate the presence of both deep and shallow carrier traps. The shallow defects which are situated at about 20meV below the conduction band dope the semiconductor leading to a substantial enhancement of the photoluminescence quantum yield in spite of carrier trapping. On the other hand, at high excitation densities relevant for lasing, we observe a highly correlated regime of photo-carriers. Such a coulomb correlation between the carriers leads to the suppression of non-radiative Auger recombination assisting the lasing action. Furthermore, we demonstrate that colloidal nanocrystals [4] represent the possibility of achieving a defect-free system, suffering from non-radiative quenching only due to sub-picosecond Auger like interactions at high excitation density.

We also investigate the origin of the peculiar broad-band photoluminescence exhibited by a selected group of two dimensional hybrid perovskites[2]. we show that the structural distortions of the perovskite lattice can determine the defectivity of the material by modulating the defect formation energies. By selecting and comparing two archetype systems, a <100> and <110>-oriented iodide-based 2D perovskite, we find that only the latter, subject to larger deformation of the Pb-X bond length and X-Pb-X bond angles, sees the formation of VF color centers, whose radiative decay ultimately leads to broadened PL. These findings highlight the importance of lattice strain-engineering to control the optoelectronic properties of this class of soft materials. By correlating the fabrication conditions to the non-radiative loss channels, this work provides essential guidelines for material engineering towards better optoelectronic device performances.

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[2] Cortecchia et al., J. Am. Chem. Soc., 139, 39 (2017).

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