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Perovskites photophysics: Half-organic, half-inorganic and a quarter of magic

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Hybrid organic-inorganic metal halide perovskites represent a remarkable success story in recent materials science applied to optoelectronic devices, thanks to the demonstrations of solution- process solar cells with conversion efficiencies in excess of 20% and very promising LEDs. The peculiarities of perovskites are thought to stem from a blend of organic materials features, like easy fabrication and bandgap tuneability, with inorganic semiconductor properties, particularly large carrier mobilities. We will show however that concerning excited state photophysics, hybrid perovskites are a unique class of materials. Ultrafast spectroscopy demonstrates that, unlike organics, perovskites are free carrier seminconductors: the prevalent excited state species are free electrons and holes in all conditions relevant for device operation, without noticeable presence of bound excitons. As a consequence, radiative efficiency increases with the excited-state density, approaching unity at high excitation when optical gain and lasing are observed. The exciton binding energy is however an important parameter, as it turns out to be larger than in most III-V inorganic semiconductors, generating an excitonic correlation strong enough to boost optical absorption and emission close to the bandgap; furthermore excitons become favoured over free carriers at low temperatures and high excitation levels. In spite of the large trap concentrations in solution-process perovskite films, optical excitations can be long-lived and radiative recombination efficient. we explore the recombination processes and demonstrate an optical technique to measure the ideality factor without any current flowing through the film, identifying and distinguishing recombination in the bulk and at each of the interfaces selectively. A picture emerges of selective traps creating unbalanced free electron and hole populations, a feature that appears to be universally shared by perovskite materials with various compositions and fabrication routes. Prospects of widespread perovskite optoelectronics are contingent on the ability to exploit their unique photophysics. As it turns out, perovskite materials may be not only a cheaper or better performing alternative to established materials, but able to perform qualitative different functions, such as vectorial charge trasnsport or density-dependent charge separation and recombination.

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