Hot, bound, and defect states – exploring the rich photophysics of two-dimensional perovskites

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Two-dimensional metal halide perovskites (2D HaPs) are exciting materials for both fundamental and applied research. Typically, these compounds consist of alternating slabs of inorganic metal-halide octahedra and organic spacer cations. This low-dimensional structure confines charge carriers and gives rise to a large exciton binding energy in excess of 100 meV and subsequently high luminescence yields. In addition, easy pathways for chemical tailoring allows for accessing a broad spectral range of emission rendering 2D HaPs attractive candidates for applications in lighting and display technology. Given their large exciton binding energy, these compounds, 2D HaPs are furthermore a rich playground for fundamental studies on excitonic physics – biexcitons, hot excitons, and bound excitons have all been reported.

In my presentation I shall focus on two pressing topics in 2D HaP research. On the one hand, I resolve a controversy surrounding often observed broad luminescence bands from some of these compounds. Such bands are prime candidates for the generation of white light and their origin is generally attributed to strong deformation of the perovskite crystal lattice in its excited state, so-called self-trapped excitons. In contrast, I shall present compelling evidence based on optical microscopy and spectroscopy studies, that broad band luminescence in these compounds actually arises from defect states.

In the second part of my talk, I shall consider the intricate sub-structure of the band edge photoluminescence in a number of 2D HaPs. The nature of emitting states – whether there are free, bound or dark excitons are involved – as well as their interaction with lattice vibrations is currently a vivid field of research. Many studies here focus on prototypical lead iodide-based compounds with a variation of organic spacers. I shall show how targeted compositional variation of the metal and halide ions allows for drawing stronger conclusions about the roles of exciton-phonon coupling, carrier cooling and the nature of emitting states.

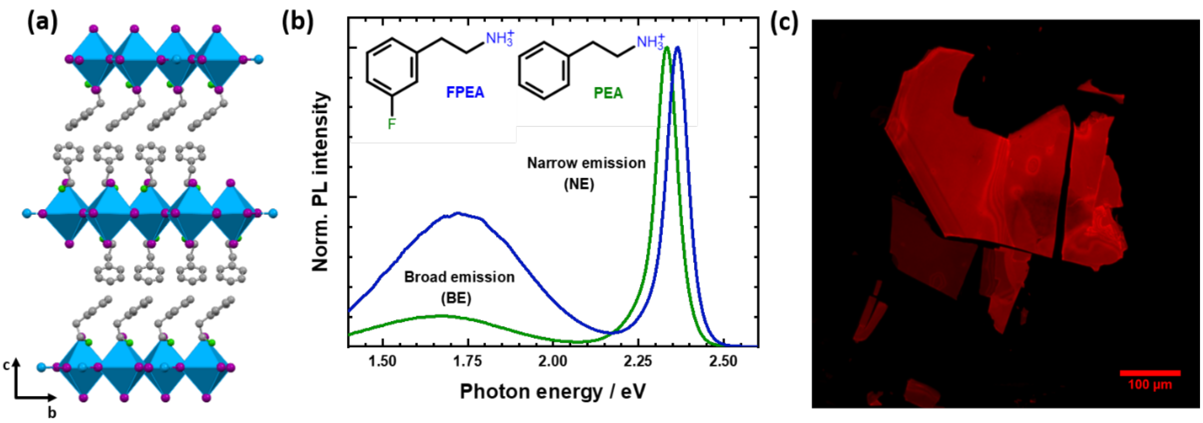

Figure 1: Crystal structure of a 2D HaP (PEA_2PbI_4) highlighting the alternating slabs of inorganic metal halide octahedra and organic spacer (a). Photoluminescence spectra of two compounds that exhibit both a pronounced free exciton emission and a broad luminescence band (b). Confocal PL map of a 2D HaP single crystal detected in the region of the broad emission band (c).