The Impact of Reabsorption on the Emission Spectra and Recombination Dynamics of Hybrid Perovskite Single Crystals

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Understanding the surface properties of hybrid perovskite is crucial to improve the efficiency of devices. Different studies suggest the existence of important differences between the surface and bulk photophysical properties of hybrid perovskites.1,2 Recently, an increase of the optical band gap of the surface layer relative to the bulk has been suggested for CH₃NH₃PbBr₃ single crystals, due to ion migration.2 Such effect would have a detrimental impact on device performance.

In this study, we have investigated the surface and bulk properties of CH₃NH₃PbBr₃ single crystals with a combination of cathodoluminescence (CL), steady-state and time-resolved photoluminescence (PL) spectroscopy. Firstly, depth-resolved CL has been used to probe the near surface region on depth ranging from a few nanometers to several micrometers. Secondly, we have studied the transmitted PL through different thicknesses. In both cases, experimental emission spectra were compared with simulated spectra, taking into account reabsorption effect. The results reveal the strong impact of reabsorption on the emission of hybrid perovskites. Reabsorption effect explains mainly the large variation of the emission spectra reported for hybrid perovskite single crystals, as well as the apparent differences between bulk and surface properties. In addition, we show that hybrid perovskite single crystals, even with millimeter size, are partially transparent to their own luminescence. The transmitted PL presents a long rising time and a lengthening of its decay due to photon recycling and light-trapping.3

Figure 1: CL spectrum at 2kV (green), transmitted PL through a 600 µm thickness (orange) and absorption coefficient (black) of CH₃NH₃PbBr₃ single crystals.4


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