

Maria Antonietta Loi

Zernike Institute for Advanced Materials, University of Groningen
Netherlands

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Understanding surface passivation in methylammonium lead tribromide single crystals

The surface of hybrid perovskites plays a crucial role in the performance and stability of optoelectronic devices, as it strongly influences the recombination rate of excited charge carriers. Recently, it has been reported that molecular ligands such as benzylamine are capable of significantly reducing the surface trap state density in thin films. Here I will report on the mechanism that governs the surface passivation of hybrid perovskites by benzylamine. To this end, we developed a versatile approach to investigate the influence of benzylamine passivation on the well-defined crystal surface of freshly cleaved methylammonium lead tribromide single crystals. We show that benzylamine is capable of permanently passivating surface trap states in these single crystals, resulting in enhanced photoluminescence intensities and charge carrier lifetimes. Additionally, we show that exposure of the perovskite surface to benzylamine leads to replacement of the methylammonium cations by benzylammonium, thereby creating a thermodynamically more stable two-dimensional perovskite (BA)₂PbBr₄ on the surface of the 3D crystal. This conversion from a 3D to 2D perovskite drives an anisotropic etching of the crystal surface, with the {100} planes being most prone to etching.