A new photophysics for 2D and 3D lead halide perovskites: Polaron plasma in equilibrium with bright excitons

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Rapid advances in perovskite photovoltaics have produced efficient solar cells, with stability and duration improving thanks to variations in materials composition, including the use of layered 2D perovskites. A major reason for the success of perovskite photovoltaics is the presence of free carriers as majority optical excitations in 3D materials at room temperature. On the other hand, the current understanding is that in 2D perovskites or at cryogenic temperatures insulating bound excitons form, which need to be split in solar cells and are not beneficial to photoconversion. Here we apply a tandem spectroscopy technique that combines ultrafast photoluminescence and differential transmission to demonstrate a plasma of unbound charge carriers in chemical equilibrium with a minority phase of light-emitting excitons, even in 2D perovskites and at cryogenic temperatures. We validate the technique with 3D perovskites and investigate 2D compounds basded on both Pb and Sn as metal cation. The underlying photophysics is interpreted as formation of large polarons, charge carriers coupled to lattice deformations, in place of excitons. A conductive polaron plasma foresees novel mechanisms for LEDs and lasers, as well as a prominent role for 2D perovskites in photovoltaics.