Small Polarons in 2D Perovskites

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We demonstrate that white light luminescence in two-dimensional (2D) perovskites stems from photoinduced formation of small polarons confined at specific sites of the inorganic framework in the form of self-trapped electrons and holes. We discuss their application in white light emitting devices and X-ray scintillators.

2D perovskites are a class of layered materials consisting of alternating organic and inorganic sheets. The resulting structure behaves like a self-assembled multiple quantum well, where charge carriers are confined within the inorganic layers and the exciton binding energy can increase up to 300 meV. In certain cases, strong excitonic characteristics lead to unusually broad, highly Stoke shifted luminescence.

Here we study the emissive properties of the 2D white light emitting perovskites (EDBE)PbX_4 (where X=Cl, Br) by steady state and time resolved spectroscopy), which reveal the formation of multiple photoinduced colour centres with wide energy distribution (Fig. 1a,b). Density functional simulations of perturbed crystal structures and large perovskite molecular clusters have allowed identifying the origin of the emissive centres, which are associated to the ultrafast formation of self-trapped electrons (STEL) Pb_2^{3+} and holes (STH) X_2^-, Pb^{3+} at specific inorganic lattice sites (Fig. 1c).[1, 2, 3]

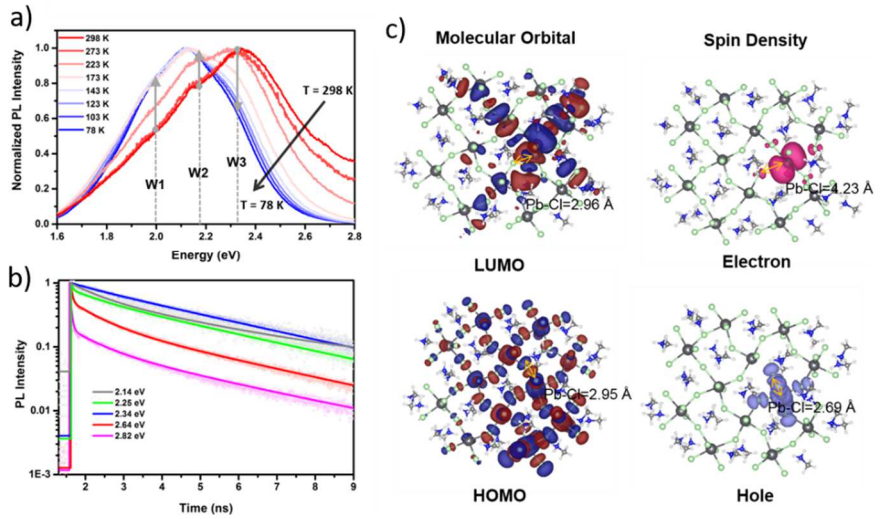

Figure 1| a) Temperature-dependent steady state photoluminescence and b) spectral dependence of time resolved photoluminescence (TRPL) under excitation energy E_{exc}=3.26 eV of (EDBE)$PbBr_4$. c) Charge density of frontier molecular orbitals and spin density of $(EDBE)_{32}Pb_{12}Cl_{56}$ cluster showing the formation of self-trapped holes and electrons.

The large Stokes shift and exciton binding energy of 2D perovskites provide unique advantages for light emitting applications by substantially reducing luminescence self-absorption and thermal quenching effects. For instance, X-ray excitation of (EDBE)$PbCl_4$ results in brighter scintillation compared to standard 3D perovskites such as $MAPbX_3$, which may find application in large-area and low-cost X-ray detectors for medical, security and scientific applications.[4] Broadband luminescence and low self-absorption are also particularly appealing for white light emitting diodes and solid state lighting.

References:

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