

## **ABSTRACT**

Halide perovskite nanocrystals (NCs) are potentially attractive for optoelectronic devices, due to their fascinating luminescent and light harvesting properties and are of the form  $\text{CsPbX}_3$  ( $X = \text{Cl, Br and I}$ ). Typically,  $\text{CsPbI}_3$  has light absorption in the visible range of the electromagnetic spectrum, which is a useful feature for fabricating optoelectronic devices. However, these materials are very prone to external perturbations like moisture, oxygen, applied bias during device measurements, laser or X-ray irradiation due to the transformation from most stable cubic phase to the yellow non-perovskite phase. So, their structural instability is still a pressing issue. Doping transition metals is a promising and inexpensive strategy to improve the stability of perovskites. This thesis explores the impact of doping suitable elements like Cu, Mn and Zn on the structural and optoelectronic behaviour of  $\text{CsPbI}_3$ , with an emphasis on electron-hole recombination, optoelectronic and phase stability. Varying fractions of Cu, Mn and Zn were doped in the as-synthesized pristine  $\text{CsPbI}_3$  and a marked enhancement in both the structural stability and optical properties of  $\text{CsPbI}_3$  was observed. From temperature-dependent photoluminescence studies, we found that an optimal doping amount of 3.7% Cu in  $\text{CsPbI}_3$  increases the fraction of cubic phase in  $\text{CsPbI}_3$  with minimization of intermediate transition states, reduced defect density and increased electron beam resilience up to a critical electron dose of  $\sim 1.68 \times 10^6 \text{ e}^-/\text{\AA}^2$ . Light emitting diodes based on the optimally doped  $\text{CsPbI}_3$  demonstrated superior performance, including a reduced turn-on voltage of 2.6 V, increases current injection efficiency, 400% increase in electroluminescence intensity and a six-fold boost in operational stability compared to pristine  $\text{CsPbI}_3$  devices. The formation energy of the systems, orbital-resolved band structure diagrams and partial density of states computed using Density Functional Theory support these results. We also investigated the hot carrier dynamics of doped  $\text{CsPbI}_3$  NCs using ultrafast transient absorption spectroscopy. Interestingly, we observed a larger bi-exciton shift in Cu-doped  $\text{CsPbI}_3$ , resulting in longer thermalization time, higher hot carrier temperature and bandgap renormalization as compared to undoped  $\text{CsPbI}_3$  which highlights the possibility of enhancing the hot carrier dynamics via dopant engineering in perovskite NCs. The defects in perovskites were also exploited to fabricate synaptic devices for neuromorphic computing. Doping Mn increased the synaptic strength. This caused high paired pulse depression values of  $\sim 180\%$  and high charge retention time in synaptic devices, with reduction in the energy consumed per spiking event based on this doped material. Introduction of a ZnO layer reversed the synaptic polarity. This improved device structure was used to demonstrate associative brain functions such as learning-forgetting and Pavlovian learning.

Thus, dopant engineering is pivotal for advancing the commercial viability of perovskites in next-generation optoelectronics.

**Keywords:** *perovskites, transmission electron microscopy, density functional theory, carrier dynamics, light emitting diodes, synaptic devices*