Nano- and single-crystals of lead halide perovskites: from bright light emission to hard radiation detection

M. V. Kovalenko$^{1,2}$

$^1$ETH Zürich, Department of Chemistry and Applied Biosciences, CH-8093, Zurich, Switzerland, $^2$Empa-Swiss Federal Laboratories for Materials Science and Technology, CH-8600, Dübendorf, Switzerland

Chemically synthesized inorganic nanocrystals (NCs) are considered to be promising building blocks for a broad spectrum of applications including electronic, thermoelectric, and photovoltaic devices. We have synthesized monodisperse colloidal nanocubes (4-15nm edge lengths) of fully inorganic cesium lead halide perovskites (CsPbX$_3$, X=Cl, Br, and I or mixed halide systems Cl/Br and Br/I) using inexpensive commercial precursors [1]. Their bandgap energies and emission spectra are readily tunable over the entire visible spectral region of 410-700nm. The photoluminescence of CsPbX$_3$ NCs is characterized by narrow emission linewidths of 12-42nm, wide color gamut covering up to 140% of the NTSC color standard, high quantum yields of up to 90% and radiative lifetimes in the range of 4-29ns. Identical synthesis methodology is perfectly suited also for hybrid perovskites CH$_3$NH$_3$PbX$_3$ [2].

Post-synthetic chemical transformations of colloidal NCs, such as ion-exchange reactions, provide an avenue to compositional fine tuning or to otherwise inaccessible materials and morphologies. While cation-exchange is facile and commonplace, anion-exchange reactions have not received substantial deployment. Here we present fast, low-temperature, deliberately partial or complete anion-exchange in CsPbX$_3$ NCs [3]. By adjusting the halide ratios in the colloidal NC solution, the bright photoluminescence can be tuned over the entire visible spectral region (410-700 nm). Furthermore, fast inter-NC anion-exchange is demonstrated as well, leading to uniform CsPb(Cl/Br)$_3$ or CsPb(Br/I)$_3$ compositions simply by mixing CsPbCl$_3$, CsPbBr$_3$ and CsPbI$_3$ NCs in appropriate ratios.

We also present low-threshold amplified spontaneous emission and lasing from CsPbX$_3$ NCs [4]. We find that room-temperature optical amplification can be obtained in the entire visible spectral range (440-700nm) with low pump thresholds down to 5±1 µJ cm$^{-2}$ and high values of modal net gain of at least 450±30cm$^{-1}$. Two kinds of lasing modes are successfully realized: whispering gallery mode lasing using silica microspheres as high-finesse resonators, conformally coated with CsPbX$_3$ NCs, and random lasing in films of CsPbX$_3$ NCs.

Here we also demonstrate that 0.5-1 centimeter large, solution-grown single crystals of MAPbI$_3$ can serve as inexpensive, operating at ambient temperatures solid-state gamma detectors (e.g. for direct sensing of photons with energies as high as mega-electron-volts, MeV) [5]. Such possibility arises from extremely high room-temperature mobility($\mu$)-lifetime($t$) product of $10^{-2}$ cm$^2$V$^{-1}$s$^{-1}$, low dark carrier density $10^9$ - $10^{11}$cm$^{-3}$ and low density of charge traps $3 \times 10^{10}$ cm$^{-3}$, and high absorptivity of hard radiation by lead and iodine atoms.