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Understanding the role of intra-gap electronic levels in scintillating lead halide perovskite nanocrystals towards effective radiation detection schemes

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Lead halide perovskites (LHP) are rapidly emerging as efficient, low-cost, solution-processable scintillators for radiation detection. Most importantly, LHP can be embedded in suitable polymeric hosts to create composite materials and produce fast, efficient, and more sensitive detectors in a cost-effective way, to meet the specific demands of all up-to-date technological and medical applications. Indeed, when embedded in polymeric matrices, LHP nanocrystals favor the enhancement of the interaction cross-section with ionizing radiation, thanks to their high atomic number, and retain exceptional levels of radiation hardness. The implementation of this novel class of scintillating composites is imperative and aims at the achievements of scintillators with improved performances. This goal is essentially linked to the fundamental understanding of the correlation between the physical-chemical properties and the luminescence features and to the comprehension of the scintillation mechanism in nanosystems, from the primary interaction with the ionizing radiation, through energy transfer and trapping processes, to the emission of light.

An essential stage in the scintillation process is the transport of free carriers generated upon the interaction between ionizing radiation and the scintillating material: it is often largely affected by the presence of trapping sites, which can capture migrating charge carriers and either delay their radiative recombination or decrease the overall scintillation efficiency, according to the characteristics of the traps involved. Therefore, carrier trapping is arguably the most critical limitation to the scintillation performance; nonetheless, no clear picture of the trapping and detrapping mechanisms to/from shallow and deep trap states involved in the scintillation process has been reported to date, as well as on the role of the material dimensionality.

We addressed this issue by performing a comprehensive study using temperature-dependent radioluminescence and photoluminescence measurements side-by-side to wavelength-resolved thermally-stimulated luminescence (TSL) and afterglow experiments on $CsPbBr_3$ with increasing dimensionality, namely nanocubes, nanowires, nanosheets, and bulk crystals. All systems are found to be affected by shallow defects resulting in delayed intragap emission following detrapping via a-thermal tunneling. TSL further reveals the existence of additional temperature-activated detrapping pathways from deeper trap states, whose effect grows with the material dimensionality, becoming the dominant process in bulk crystals.

In addition, we prove that $CsPbBr_3$ nanocrystals can be effectively embedded into polymethylmethacrylate matrix to obtain high optical quality flexible and smooth scintillating nanocomposites, whose defectiveness resembles that of LHP bulk crystals, featuring isolated energetically deep defect states that trap carriers which, upon heating, recombine in a specific intragap emission center, as revealed by TSL measurements.

These results highlight that, compared to massive solids where the suppression of both deep and shallow defects is critical, low dimensional nanostructures are more promising active materials for LHP scintillators, provided that their integration in functional devices meets efficient surface engineering.

The effectiveness of this investigation approach coupling scintillation and TSL measurements, traditionally exploited only for classical single component bulk scintillators, is therefore demonstrated also for nanomaterials.

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