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Temperature-dependent photoluminescence of CsPbX₃ nanocrystal films

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Abstract: Temperature-dependent photoluminescence (PL) properties of all-inorganic perovskite CsPbX₃ (X=Cl, Br, I, or their mixtures) nanocrystal (NC) films are studied by use of steady-state and time-resolved PL spectroscopies. It is confirmed that the PL intensity of the NC films decreases rapidly with increasing temperature below 300 K and is nearly invariant till 370 K due to thermal quenching and degradation, respectively. With increasing temperature, photo energies of linewidth and emission peak become larger due to stronger exciton-phonon coupling. It is found that temperature-dependent PL is composed of a bandedge excitonic state and trapping state emission and produces the observation of biexponential kinetics. The short-lived emission is due to band-edge exciton recombination, while the component with long-lived lifetime is ascribed to trapping state, which arises from recombination in NC that has a photoinduced trapped pathway and a temporally resolved peak shift. Besides organic-inorganic perovskite hybrid NCs, trapping state also exists in all-inorganic CsPbX₃ NCs, even though PL spectrum has high PL quantum yield and narrow emission linewidths.

Keywords: CsPbX₃, nanocrystals, trapping state, band-edge exciton, photoluminescence

1. Introduction

In the past few years it has witnessed important progress demonstrating excellent optoelectronic characteristics of organic-inorganic lead-halide-based perovskite in the form of thin films, nanocrystals (NCs) and bulk single crystals, which is emerging as one of the most promising materials for solution-processable photovoltaic technology [1, 2]. Recently colloidal NCs of all-inorganic lead halide perovskite have attracted much attention for applications in light emitting diode (LED) [3], optically pumped lasers [4] and quantum dots displays [5]. Highly luminescent colloidal CsPbX₃ (X=Cl, Br, I, or their mixtures) NCs are synthesized using a high-temperature hot-injection approach or anion exchange reactions [6]. By adjusting composition and stoichiometry of halide element in the colloidal NC solution, the bright photoluminescence (PL) can be tuned over the entire visible spectral region while maintaining PL quantum yield (PLQY) of 60-80% and narrow emission linewidths of 10-40 nm [7].

Compared with conventional Cd-based NCs, the CsPbX₃ NCs appear to be largely free of mid-gap trap states, which enable their advantages over the Cd chalcogenide working in the blue-green spectral region of 410-530 nm [6]. To gain a deeper insight into the photophysical processes occurring in the all-inorganic CsPbX₃ NCs, it is worth exploring them at low temperatures where the additional complexity induced by thermal effects is minimized. In organic-inorganic hybrid perovskite NCs, particularly in well-studied CH₃NH₃PbBr₃ NCs [8, 9], double PL emission peaks can be observed in temperature-dependent steady-state PL measurement, the structural phase transition from tetragonal to orthorhombic exists at low temperature of 150 K for CH₃NH₃PbI_{3-x}Cl_x [10], 160 K for CH₃NH₃PbBr₃ NCs [11], 150 K

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