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Article

Surface Ligand Engineering to Modulate the Synthesis of DJ-Type 2D Sn-Based Perovskites for Light-Emitting Diodes

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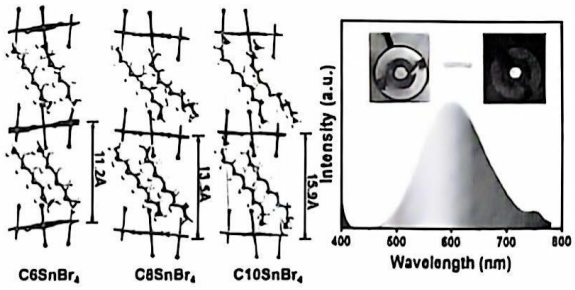
ABSTRACT: The advancement of novel lead-free and stable metal halide perovskites is crucial in addressing the environmental risks associated with lead-based perovskites and broadening their range of applications. Two-dimensional (2D) Sn-based perovskites have been increasingly recognized for their outstanding optoelectronic properties and notable stability. In this study, we have successfully synthesized three variants of Dion–Jacobson (DJ)-type 2D Sn-based perovskites ($H_3NC_nH_{2n}NH_3$)$SnBr_4$ through surface ligand modification using 1,6-diaminohexane (C6), 1,8-octanediamine (C8), and 1,10-decanediamine (C10). $C6SnBr_4$ exhibits an exceptionally high absolute photoluminescence quantum yield (PLQY) of up to 95%. Analysis of low-temperature PL spectra sheds light on the luminescence mechanism of $C6SnBr_4$, emphasizing the interaction between band-edge emission and self-trapped state emission. Density functional theory (DFT) calculations reveal a bandgap (E_g) of 2.07 eV for $C6SnBr_4$, consistent with experimental findings. Furthermore, the monochromatic phosphor-converted light-emitting diode (LED) incorporating $C6SnBr_4$ displays a correlated color temperature (CCT) of 2483 K, emitting a vibrant yellow-orange hue with a color rendering index (R_a) of 47.7, and CIE color coordinates of (0.514, 0.471). These findings offer valuable insights for enhancing the absolute PLQY and stability of DJ-type 2D Sn-based perovskites.



1. INTRODUCTION

Metal halide perovskites (MHPs) are widely embraced for their outstanding optoelectronic properties, finding applications in photovoltaics,[1,2] light-emitting diodes (LEDs),[3,4] photodetectors,[5] fluorescence imaging,[6] and photocatalysis data memories et al.[7,8] However, the environmental concerns associated with Pb-based halide perovskites have underscored the need for research on Pb-free alternatives.[9−12] Among these, Sn-based halide perovskites have gained interest by reason for their similarities with Pb in various aspects.[13−16] Despite their promising features, MHPs face challenges in stability due to their inherent ionic structural properties, rendering them vulnerable to external factors like solvents, ultraviolet (UV) rays, temperature, humidity, and electric fields. This limitation significantly impacts the practical utilization of MHPs.[17,18]

The reason for the growing interest in two-dimensional (2D) MHPs is their exceptional water and thermal stability.[13,19,20] The structure of 2D MHPs maintains the fundamental perovskite structure, where octahedral units made of metal halides are linked through shared corners. Moreover, long-chain organic amine cations connect the inorganic layers comprised of octahedra. The inclusion of hydrophobic organic long-chain amine ligands provides 2D MHPs with resistance to oxygen and moisture in the atmosphere, enhancing their long-term stability.[20] These organic long-chain amine cations are divided into monoamine and diamine categories, giving rise to two types of 2D MHPs: Ruddlesden–Popper (RP)-type and Dion–Jacobson (DJ)-type, respectively.[21−23] In RP-type 2D MHPs, monoamine organic long chains connect adjacent inorganic layers through van der Waals forces, impeding electron transfer. Conversely, in DJ-type 2D perovskites, long organic chains of diamines link neighboring inorganic layers via hydrogen bonding between amine ions and octahedra at both ends. This arrangement enhances structural stability and promotes electron transfer in DJ-type 2D perovskites.[23−29]

A growing passion has been observed in recent years in DJ-type 2D Sn-based perovskites. Rogach et al. utilized a room

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