Oxide Encapsulation of $CsPbBr_3$ Nanocrystals to Enhance the Optical Efficiency and PLQY for LEDs Through Transition Dipole Alignments

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Abstract

The efficiency of perovskite light-emitting diodes (PeLEDs) is often hindered by the degradation of quantum dots (QDs) due to interactions with air, moisture, and substrate. To address this, we demonstrate that encapsulating $CsPbBr_3$ nanocrystals with a silica (SiO_2) layer significantly enhances their stability and optical performance. Silica-encapsulated $CsPbBr_3@SiO_2$ retained 76.8% of its initial photoluminescence (PL) intensity after two weeks of exposure to light and moisture, compared to only 34% for uncoated $CsPbBr_3$. Furthermore, oxide encapsulation improves light extraction by reducing total internal reflection within the active layer, directing emissions towards the escape cone. The transition dipole moment (TDM) angle for $CsPbBr_3@SiO_2$ was measured to be ~32°, indicating a tendency towards isotropic emission, which enhances outcoupling efficiency. These findings suggest that oxide encapsulation is a promising strategy for improving the operational stability and optical efficiency of PeLEDs.

Author Keywords

PLQY; Perovskite; EQE; Lifetime; SiO_2 encapsulation

Introduction

Colloidally synthesized perovskite nanocrystals ($CsPbX_3$) exhibit exceptional optical properties, including near-unity photoluminescence quantum yield (PLQY) and highly directional emission [1][2]. In recent years, the external quantum efficiency (EQE) of perovskite-based LEDs has significantly improved from 1% to 20% [3]. However, further enhancement in EQE can be achieved by optimizing light emission alignment.

Despite their remarkable photophysical properties and high PLQY, cesium lead halide perovskites are highly susceptible to degradation when exposed to water, oxygen, and light. Water absorption leads to the formation of non-photoluminescent phases such as $Cs_4PbBr_6{\cdot}2H_2O$ and $CsPb_2Br_5$, reducing their optical performance [4]. This degradation occurs due to water and oxygen adsorption at the perovskite surface—water molecules interact with both CsBr- and PbBr-terminated surfaces, whereas oxygen preferentially binds to CsBr-terminated surfaces, accelerating material degradation [5][6][7]. The degradation reaction is represented as

$$7CsPbBr_3 + 2H_2O \rightleftarrows Cs_4PbBr_6{\cdot}2H_2O + 3CsPb_2Br_5 \quad (1)$$

A promising approach to mitigate degradation involves encapsulating $CsPbBr_3$ nanocrystals within a stable oxide or molecular shell. Encapsulation within Al_2O_3 and SiO_2 matrices has been shown to significantly enhance the stability of perovskite nanocrystals. For instance, $CsPbBr_3@SiO_2$ core-shell nanoparticles synthesized by Qixuan Zhong et al. retained stability for over seven days in water while maintaining a high PLQY of 90%. During the synthesis process of silica encapsulation silica precursors Tetramethyl orthosilicate (TMOS) was added into $CsPbBr_3$ dissolved in DMF. TMOS hydrolyses on the surface of perovskites in the presence of water to form SiO_2 layer on $CsPbBr_3$ nc's the reaction is stopped by adding in ammonia (NH_3) [12]. This encapsulation provides a physical barrier, preventing environmental degradation while also improving exciton recombination, charge transfer, and suppressing non-radiative Auger recombination [8][9]. The optical and electronic properties of such core-shell structures can be finely tuned by selecting appropriate core and shell materials [10][11].

Furthermore, modifying the perovskite nanocrystal surface alters the transition dipole moment (TDM) orientation, which is sensitive to the local dielectric environment. The anisotropic electronic environment created by placing perovskites on a glass substrate (air-perovskite above, perovskite-glass below) increases the TDM angle [1][2]. Introducing an additional surface layer counterbalances the substrate's influence, reducing the TDM angle and enhancing anisotropic light emission. Materials with work functions similar to the substrate can induce a comparable charge reconfiguration, creating an opposing field to the substrate-induced dipole distortion [12]. For instance, Jurrow et al. demonstrated that coating perovskites with an Al_2O_3 layer reduced the dipole angle by 50%, from 29 ± 1° to 14°, due to suppression of substrate-induced vertical dipoles [13].

In this work, we demonstrate that such surface modifications can be achieved colloidally, eliminating the need for vacuum-based deposition processes. By leveraging oxide encapsulation and precise control over the dielectric environment, we present a scalable strategy for stabilizing perovskite nanocrystals while optimizing their emission properties.

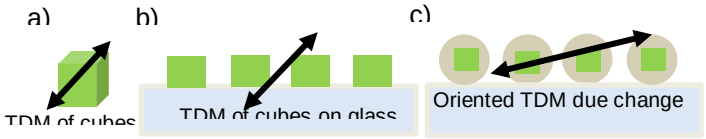

Figure 1 a, b) Angular emission of TDM of bare cubes and cubes on glass substrate c) Angular emission of TDM of $CsPbBr_3@SiO_2$ on glass substrate.

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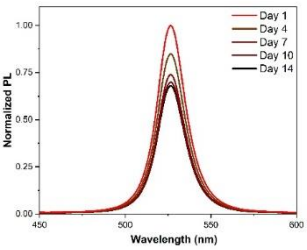
Introducing an additional surface layer on top of perovskites through partial shelling can effectively counterbalance the substrate's influence, thereby further reducing the transition dipole moment (TDM) angle relative to the substrate. By selecting a material with a work function similar to that of the substrate, a comparable charge reconfiguration is induced on the perovskite surface. This reconfiguration generates an electric field that opposes the substrate-induced dipole distortion, leading to improved emission directionality and enhanced optical performance [13][8].

Results

To evaluate the degradation rate, we investigated the stability of $CsPbBr_3@SiO_2$ nanocrystals by monitoring their photoluminescence (PL) intensity over a period of two weeks. The $CsPbBr_3@SiO_2$ particles were synthesized via a sol-gel reaction mechanism. As shown in Figure 2c, the encapsulated $CsPbBr_3@SiO_2$ particles exhibit an emission peak at 528 nm and an absorption peak at 516 nm, which closely match the PL emission range of $CsPbBr_3$ nano cubes (515–530 nm). This consistency indicates that SiO_2 encapsulation does not induce a shift in the PL peak, confirming that silica does not alter the band alignment of the perovskite.

To assess their environmental stability, both uncoated $CsPbBr_3$ cubes and encapsulated $CsPbBr_3@SiO_2$ were exposed to air, moisture, and light for 14 days. As seen in Figure 2b, the PL intensity of uncoated $CsPbBr_3$ cubes declined by more than 75% of its initial value, whereas the encapsulated $CsPbBr_3@SiO_2$ particles exhibited only a 34% reduction. This significant improvement in stability suggests that the SiO_2 shell effectively protects $CsPbBr_3$ from environmental degradation. Additionally, the absence of a red or blue shift in the PL emission peak confirms that SiO_2 encapsulation does not impact charge transfer within the perovskite, preserving its optoelectronic properties.



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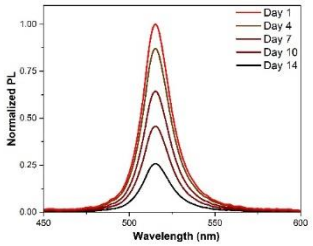

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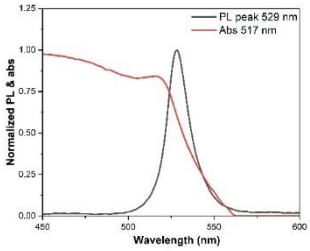
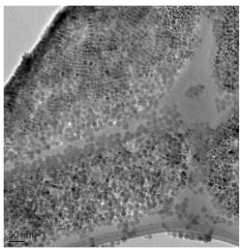


Figure 2 a) Photostability of $CsPbBr_3@SiO_2$ exposed to air and water b) Photostability of $CsPbBr_3$ cubes under UV irradiation c) Photoluminescence spectra of $CsPbBr_3@SiO_2$ nanoparticle

The TEM image of the encapsulated particles Fig 3a and 3b confirmed the presence of SiO_2 molecule. The spherical SiO_2 particles are attached on the surface of the cubic perovskites (Color Contrast the lighter particles SiO_2 and the darker one $CsPbBr_3$ cubes). In the presence of excessive silica precursor silica particles can hydrolyze in the presence of water to react to form additional SiO_2 nanoparticles. Hence controlling the concentration of the TEOS precursors and the rate of hydrolysis can determine the ratio of SiO_2 to $CsPbBr_3$ nanoparticles. The optimum ratio of TMOS concentration to $CsPbBr_3$ solution concentration was determined to be 1:400 to achieve encapsulation and lesser aggregates of SiO_2. From the TEM image in figure 3b the size of $CsPbBr_3$ and SiO_2 can be calculated using Image Js software to be around 11.5 nm and 23 nm as seen in Figure 3c and 3d.

a)



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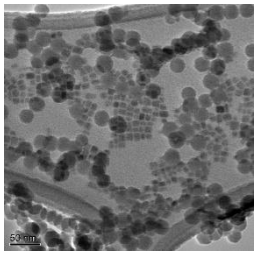

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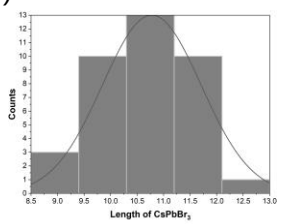

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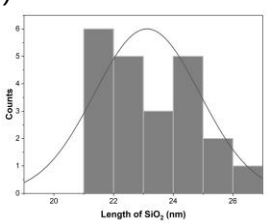

Figure 3 a,b) Transmission electron microscopy of $CsPbBr_3@SiO_2$ NC's 10x and 20x dilutions c, d) Length(nm) of $CsPbBr_3$ and SiO_2 nanoparticles measured from the TEM

The angular light emission pattern was measured using back focal imaging which results in the 2D projection of a 3D angular emission pattern (Figure 4d) emissions from the face of the device is along θ direction whereas ϕ represents light emissions along edge of the device. Figure 4c shows the horizontal and vertical cross section of the back focal plane image and the corresponding theoretical curve. Because the angular light emission from a sample is a direct function of the alignment of the transition dipole moment, one can use the light emission pattern to determine the average angle of the transition dipole moment [17][13].

The light emissions, i.e. the transition dipole orientation from perovskite nc's is a function of the local environment. Visually, there are strong differences between the films resulting from the amount of horizontal dipoles versus vertical dipoles. In figure 4a for a TDM angle of 0 ° corresponds to strong emission in the k_y direction and strong dips along the k_x direction (Figure 4 a) i.e. most of light emissions will be from the face of the LEDs. This results from the strong light emission perpendicular to the dipole and low emissions in the direction. A TDM of 90° corresponds to strong emission in the k_x direction (Figure 4 c) resulting in light emissions from the edges of the LED's. Most of the light will be emitted perpendicularly to the dipole, resulting in light emissions at a high angle and because of the polarizer, we only see light emitted along k_x. Any angle in between horizontal and vertical will be a summation of these two contributions (Figure 4 b).

From the Figure 4 d) we can observe strong dipoles in the k_x direction and from 4d) the angular emission was calculated to be at 33.2°. Unshelled $CsPbBr_3$ cubes which have an average TDM angle at 40-45°, indicating that encapsulation enhances the in-plane light emissions compared to the unshelled $CsPbBr_3$ cubes, which would help the outcoupling efficiency. To further improve the fit, I will calculate the precise value of SiO_2 and $CsPbBr_3$ by volume percentages and packing density of the particles. I hypothesize that alignment of these particles will lead to even more anisotropic emissions.

To further refine the emission profile, we have calculated the precise volume percentages and packing density of SiO_2 and $CsPbBr_3$ to estimate the refractive index of the encapsulated particles to be around 1.69 using Bruggeman analysis [18]. We hypothesize that optimizing the surface thickness and refractive index of these particles has led to accurate calculation of light emission (dipole angle) from the spin coated thin films.

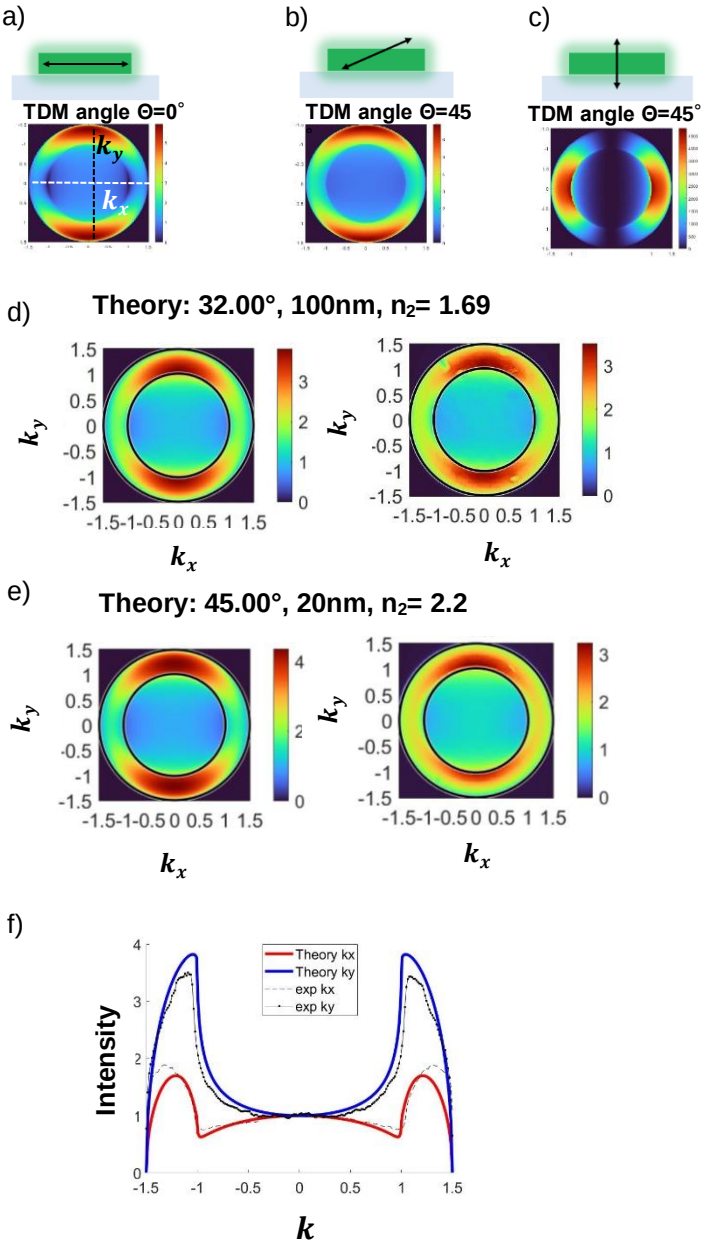

Figure 4 a, b, c) Correlation between light emission to angle of TDM 0° TDM, 45° TDM and 90° TDM respectively d) Back focal plane (BFP) image of $CsPbBr_3$@SiO_2 np's left theoretical BFP and right experimental BFP emission pattern e) Back focal plane(BFP) image of $CsPbBr_3$ cubes left theoretical BFP and right experimental BFP emission pattern f) fitting of experimental data to theory for $CsPbBr_3$@SiO_2 np's

Conclusion

We have successfully synthesized $CsPbBr_3$@SiO_2 particles through a sol gel synthesis; the PL and absorption spectra are 524 nm and 515 nm similar to that of $CsPbBr_3$. The $CsPbBr_3$ SiO_2 can be tuned by varying the halide ion i.e Br with I and Cl to achieve color tunability of this material from 441 nm to 515 nm wavelength which makes these ideal materials for

LED applications. Photo degradation of these materials improved 34% reduction of the PL intensity of bare $CsPbBr_3$ cubes to 75% for $CsPbBr_3@SiO_2$ molecule. The TDM angular emissions of the $CsPbBr_3@SiO_2$ in is around 32° the TDM angle for cubes is around 45° this proves that the refractive index (interaction of light with local environment) of the SiO_2 (Local environment) plays a role in tuning the light emissions i.e refractive index of the SiO_2 being similar to the glass substrate orients the dipoles thus the light emission from $CsPbBr_3@SiO_2$ tend towards anisotropy.

Impact of Research

We investigated the effects of silica shelling on the stability and angular light emission of perovskite nano cubes. Perovskites are highly surface-sensitive materials, meaning their physical properties are strongly influenced by the surrounding medium, such as solvents and substrates. Notably, the dipole alignment in perovskites is affected by changes in the local environment, including trapped surface charges. My observations revealed that the attachment ofSiO_2 significantly stabilizes the perovskites, slowing their rate of degradation. Partial encapsulation also influenced the average transition dipole moment (TDM) and angular light emission. Specifically, the TDM angle for bare $CsPbBr_3$ nano cubes was approximately **45°**, whereas SiO_2-encapsulated $CsPbBr_3$ exhibited TDM angles around **32°**, indicating a shift toward more parallel-oriented dipoles which would mean a higher probability of light outcoupling and higher EQE. Further studies, particularly of the photoluminescence quantum yield (PLQY) over extended time scales, will provide deeper insights into the degradation mechanisms and the long-term benefits of SiO_2 encapsulation.

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