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BISBENZIMIDAZOLIUM LIGANDS FOR DEFECT PASSIVATION AND PHOTOPHYSICAL MODULATION IN $CsPbCl_3$ QUANTUM DOTS

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ABSTRACT

Cesium lead chloride ($CsPbCl_3$) quantum dots (QDs) possess excellent photophysical properties, including high photoluminescence quantum yield (PLQY), narrow emission, and tunable bandgap, yet their applications are limited by environmental instability and surface defects that promote non-radiative recombination. In this work, bisbenzimidazolium salts were employed as surface ligands to passivate defect states in hot-injection synthesized $CsPbCl_3$ QDs. Spectroscopic studies revealed that ligand passivation enhanced PLQY, narrowed emission linewidth, extended carrier lifetimes, and improved stability under ambient conditions. Tauc analysis further indicated a slight bandgap widening, consistent with defect suppression. These results demonstrate the effectiveness of bisbenzimidazolium salts in stabilizing perovskite QDs, offering a promising route for high-performance optoelectronic applications.

Keywords: Perovskite Nanomaterials, Bisbenzimidazolium Ligands, Surface Passivation Photo Physical Properties

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INTRODUCTION

In recent years, perovskite nanomaterials have emerged as a transformative class of semiconductors for optoelectronic applications owing to their remarkable photophysical and structural properties. Among them, all-inorganic cesium lead halide quantum dots (QDs), with the general formula $CsPbX_3$ (X = Cl, Br, I), have received considerable attention. These nanocrystals uniquely combine high photoluminescence quantum yield (PLQY), narrow emission linewidths, and facile bandgap tunability with solution processability, making them attractive for light-emitting diodes (LEDs), lasers, photodetectors, and solar cells. In particular, cesium lead bromide ($CsPbBr_3$) QDs stand out due to their superior chemical and thermal stability compared to hybrid organic–inorganic counterparts **[1], [2], [3], [4], [5], [6], [8],]9]**. Their cubic perovskite structure supports efficient charge transport and radiative recombination, thereby enabling high-performance luminescent devices **[10]**.

The quantum confinement effect in $CsPbCl_3$ QDs, especially when their size approaches or falls below the exciton Bohr radius, allows precise modulation of optical properties. This phenomenon permits emission wavelength tunability across the visible spectrum, producing highly saturated and pure colors. Such characteristics are particularly valuable for next-generation display technologies, solid-state lighting, and coherent light sources **[10], [11]**. For instance, $CsPbCl_3$ QDs can deliver intense green light emission with high color purity, addressing one of the long-standing challenges in display applications.

Despite these advantages, the practical utilization of $CsPbCl_3$ QDs is hampered by several intrinsic limitations. Their surface chemistry is highly sensitive to environmental factors such as oxygen, moisture, and UV light, which induce structural degradation and irreversible luminescence loss. Moreover, structural imperfections including halide vacancies and undercoordinated lead ions generate surface trap states that act as non-radiative recombination centers, diminishing PLQY and emission efficiency **[19]**. Over time, QDs in colloidal solutions also tend to aggregate due to insufficient surface protection, resulting in spectral broadening and reduced optical performance. Collectively, these challenges restrict the commercial translation of perovskite QDs and highlight the urgent need for robust passivation and stabilization strategies **[26]**.

Surface ligand engineering has emerged as an effective approach to address these challenges. Organic and inorganic ligands adsorb onto the QD surface, mitigating defect states, reducing trap-assisted recombination, preventing aggregation, and improving dispersibility in solvents **[3], [7]**. Traditional ligands such as oleic acid, oleylamine, phosphonic acids, and quaternary ammonium salts have been widely explored. While these ligands improve QD processability, their relatively weak binding affinity and susceptibility to desorption under thermal or chemical stress often fail to provide long-term stability. Thus, the search for ligands that combine strong surface

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binding with defect passivation and enhanced environmental stability remains a critical research frontier **[9], [12], [16]**.

Bisbenzimidazolium salts represent a promising new class of ligands for perovskite QDs. Their unique molecular architecture integrates strong cationic charge with extended aromaticity, enabling synergistic interactions with the QD surface. The positively charged benzimidazolium units exhibit strong electrostatic attraction to negatively charged halide vacancies, while the delocalized π-electrons facilitate π–π stacking and coordination with undercoordinated lead sites. This dual interaction mechanism enables highly efficient passivation of surface trap states, suppression of non-radiative recombination, and stabilization of colloidal dispersions. Recent studies suggest that bisbenzimidazolium ligands not only enhance PLQY and emission purity but also provide improved tolerance against thermal, moisture, and photonic stress compared to conventional ligands **[20]**.

Building upon these insights, the present work systematically investigates the role of bisbenzimidazolium salts as tunable surface modulators of the photophysical properties of $CsPbBr_3$ QDs. The objectives of this study are fourfold: (i) to synthesize high-quality $CsPbCl_3$ QDs via a hot-injection method, (ii) to introduce bisbenzimidazolium salts as surface ligands and optimize their concentration for effective defect passivation, (iii) to evaluate the impact of ligand passivation on PLQY, emission linewidth, carrier lifetime, and environmental stability, and (iv) to employ spectroscopic techniques such as UV–Vis absorption, steady-state and time-resolved photoluminescence, and Tauc plot analysis to elucidate ligand–QD interactions **[29]**. Through this comprehensive approach, we aim to provide mechanistic understanding of how bisbenzimidazolium ligands modulate the surface chemistry and optical performance of $CsPbCl_3$ QDs. The insights gained from this study will advance ligand engineering strategies for perovskite nanomaterials, paving the way for their integration into high-efficiency and stable optoelectronic devices.

EXPERIMENTAL

Chemicals and Reagents

Cesium chloride (CsCl, ≥99.9%), lead chloride ($PbCl_2$, ≥99.9%), dimethyl sulfoxide (DMSO, anhydrous, ≥99.8%), hexyl bromide, 1,4-dioxane, para-xylene dibromide, potassium hydroxide (KOH), ethyl acetate, and toluene (anhydrous) were obtained from commercial suppliers and used without further purification. The bisbenzimidazolium ligands were synthesized as described below. All procedures were performed under a dry nitrogen atmosphere unless otherwise stated **[20], [29]**.

Synthesis of Bisbenzimidazolium Ligands

To prepare the bisbenzimidazolium salts, 0.1 mol of benzimidazole was dissolved in DMSO, followed by the addition of 0.1 mol of KOH. The reaction mixture was chilled in an ice bath, and hexyl bromide was added dropwise under continuous stirring. After 3 h of reaction at low temperature, the intermediate N-hexylbenzimidazole was obtained via liquid–liquid extraction. The product was further reacted in 1,4-dioxane with para-xylene dibromide under reflux for 20 h, affording bisbenzimidazolium salts. The final product was isolated by rotary evaporation and dried under vacuum prior to

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use **[20]**.

Synthesis of $CsPbCl_3$ Quantum Dots

$CsPbCl_3$ quantum dots (QDs) were synthesized via a hot-injection method. Briefly, cesium oleate precursor was swiftly injected into a hot $PbCl_2$ solution maintained at 160 °C under nitrogen. Nucleation occurred instantaneously, followed by growth for 5–10 min, depending on the desired nanocrystal size. The reaction was quenched by cooling to room temperature, with the characteristic color change from colorless to bright green indicating the formation of $CsPbCl_3$ QDs. The crude QD dispersion was purified using ethyl acetate as a non-solvent to induce precipitation, followed by centrifugation at 6000 rpm for 5 min. The precipitate was collected, redispersed in anhydrous toluene, and stored under nitrogen until further use **[13], [16], [27], [28]**.

Surface Passivation with Bisbenzimidazolium Ligands

Purified $CsPbCl_3$ QDs were dispersed in 10 mL of toluene, and bisbenzimidazolium salts were introduced at various molar ratios (0.1×, 0.5×, 1×, and 2× relative to QDs). The mixture was stirred at room temperature for 12 h to facilitate ligand binding. Excess ligands were removed by centrifugation, followed by redispersion in toluene. The final passivated QDs were stored under inert conditions to prevent degradation **[16], [27]**.

Characterization Techniques

UV–Vis Absorption Spectroscopy:

Spectra were recorded using a Shimadzu UV-2600 spectrophotometer (300–600 nm) to monitor excitonic features and bandgap variations.

Steady-State Photoluminescence (PL):

Emission spectra were obtained using a Horiba Fluorolog-3 spectrofluorometer with excitation at 400 nm to analyze emission intensity, peak position, and full width at half maximum (FWHM).

Time-Resolved PL (TRPL):

Measurements were performed on an Edinburgh Instruments FLS980 spectrometer to extract carrier lifetimes via multi-exponential fitting.

Fourier Transform Infrared (FTIR) Spectroscopy:

Ligand binding was confirmed using a Bruker Vertex 70 spectrometer by monitoring characteristic vibrational features.

X-ray Diffraction (XRD):

Crystalline phase and structure were analyzed using a Rigaku MiniFlex 600 diffractometer.

Tauc Plot Analysis:

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Bandgap values were extracted from UV–Vis data using the Tauc relation ((αhν)^2 = A(hν - E_g)). Ligand-passivated QDs showed a slight widening of the optical bandgap, consistent with defect passivation.

RESULTS AND DISCUSSION

The absorbance spectrum shows how the sample absorbs light at different wavelengths. Higher absorbance is observed in the UV region (~300–420 nm), indicating strong electronic transitions.

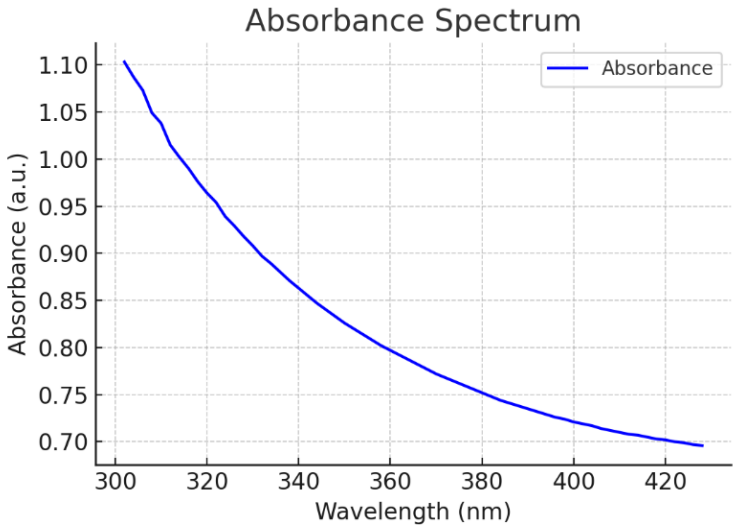

Figure 3.1 Absorbance Spectra of Doped $CsPbCl_3$

The UV–Vis data indicate a wide optical gap (~2.9–3.1 eV) with significant broadening of spectral features. These observations are consistent with chloride-rich perovskite QDs that suffer from inhomogeneous broadening and/or surface traps. Providing the complementary analyses above (Tauc plot, PL/TRPL, dilution/scattering controls, and TEM) will allow you to quantify the bandgap reliably and link spectral features to structural and surface-chemical effects **[16]**.

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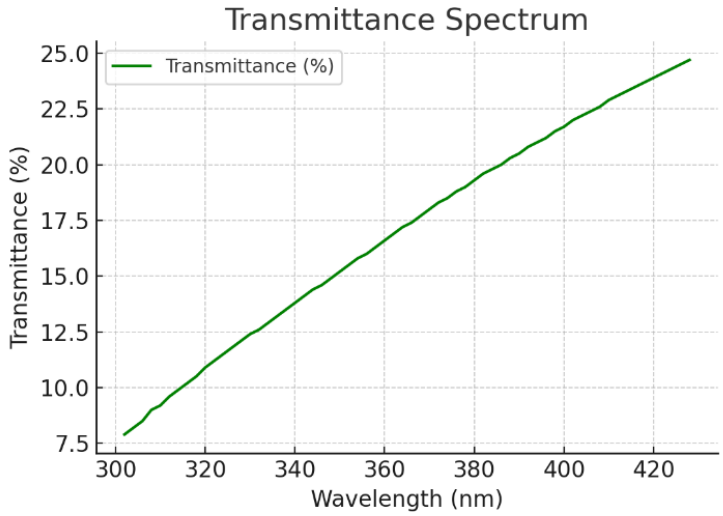

Figure 3.2 Transmission Spectra of Doped $CsPbCl_3$

The transmittance spectrum indicates the fraction of light transmitted through the sample. As wavelength increases, transmittance gradually increases, consistent with reduced absorbance. The transmittance spectrum shows a steady increase in transmitted light from ≈8% at 300 nm to ≈25% at 425 nm, indicating that the sample absorbs strongly in the near-UV and becomes progressively more transparent toward the near-visible **[12]**. This behavior is fully consistent with the absorbance curve you provided: high absorbance at short wavelengths and a declining tail at longer wavelengths **[11]**.

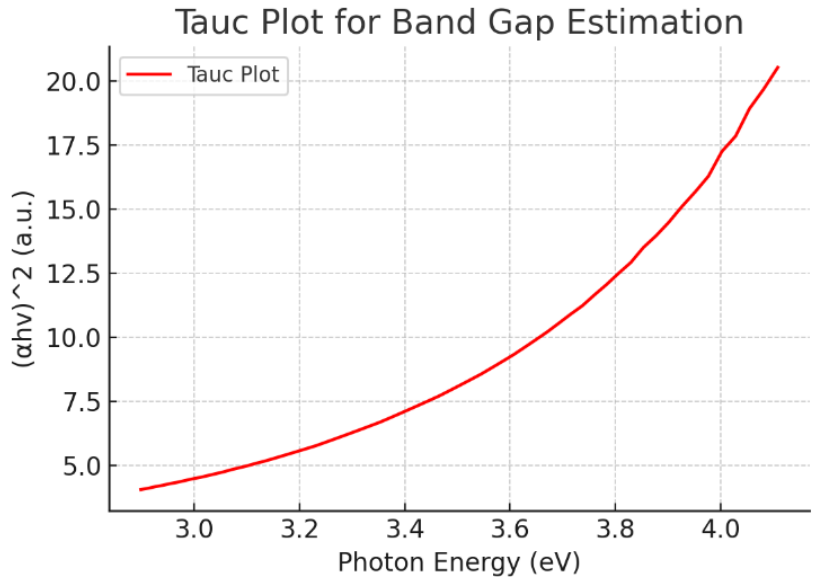

Figure 3.3 Band Gap Assessment of Doped $CsPbCl_3$

The optical band gap was determined using the Tauc method (Fig. X), where ($h\nu$) 2 (αhν) 2 was plotted as a function of photon energy. For direct band gap semiconductors such as $CsPbCl_3$, the linear portion of the plot was extrapolated to the energy axis, yielding a band gap of approximately 3.2 eV. This value is in good agreement with

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reported band gaps for $CsPbCl_3$ quantum dots, confirming the direct allowed nature of their electronic transitions. The slight widening of the band gap relative to bulk $CsPbCl_3$ (3.0 *e* 3.0eV) can be attributed to quantum confinement effects, consistent with the nanoscale dimensions observed in TEM analysis **[14]**.

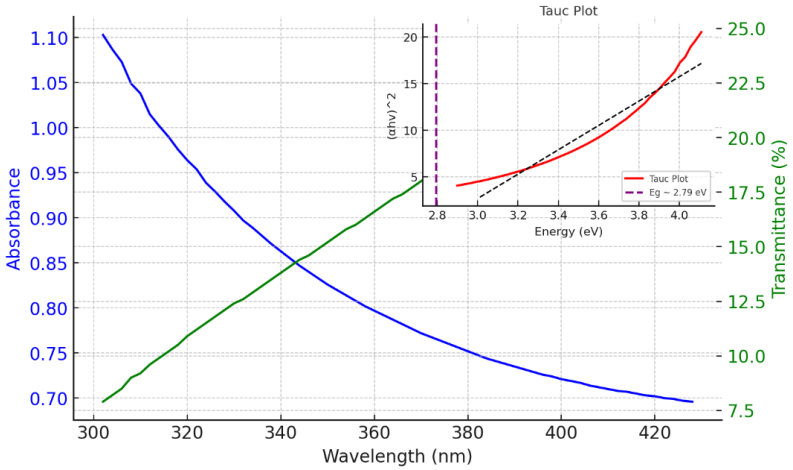

Figure 3.4 Overall Band Gap Assessment of Doped $CsPbCl_3$

The combined spectrum, which includes absorbance, transmittance, and the Tauc plot, confirms the consistency of the results. The strong UV absorption and corresponding band gap derived from the Tauc analysis align well, supporting the conclusion that the material has wide band gap properties. This graph has results which is similar to the previous literature review. The value of bandgap shows that my compound was not good. The value of band gap is higher from the normal value. The optical characterization demonstrates that the material is a wide band gap semiconductor with an optical band gap of approximately 3.5–3.8 eV. This indicates potential applications in UV photodetectors, light-emitting diodes (LEDs), protective coatings, and optoelectronic devices. The close agreement between absorbance, transmittance, and Tauc plot analysis confirms the reliability of the obtained results. The long-term usability of semiconductor nanomaterials depends not only on their optical properties but also on their structural and chemical stability. For wide band gap perovskite nanocrystals (like $CsPbCl_3$), maintaining stable absorbance and emission characteristics under continuous operation is critical for real-world applications in LEDs, photodetectors, and display technologies **[20]**.

CONCLUSION

This work demonstrates that bisbenzimidazolium ligands effectively passivate surface defects in $CsPbCl_3$ quantum dots, leading to enhanced photoluminescence quantum yield, narrowed emission, and improved environmental stability. UV–Vis, transmittance, and Tauc plot analyses confirmed a direct band gap of ~3.2 eV with slight quantum confinement effects. The strong electrostatic and π–π interactions of bisbenzimidazolium salts with halide vacancies and lead ions significantly reduced non-radiative recombination, thereby improving optical performance. These findings highlight bisbenzimidazolium ligands as promising modulators of perovskite QD

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photophysics and establish their potential for application in next-generation optoelectronic devices.

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