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Theoretical Design and Optoelectronic Analysis of Lead-Free $CsPbX_3/Cs_2SnX_6$ Core–Shell Perovskite Nanocrystals for Enhanced Stability and Charge Dynamics

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Abstract | **Original Research Article**

Due to their outstanding optical features such as high PLQY, very narrow emission and the option to change their bandgap, inorganic cesium lead halide ($CsPbX_3$, X = Cl, Br, I) nanocrystals (NCs) have gotten much recognition. Nevertheless, environmental problems and the toxicity of lead have made it hard to use these compounds in lighting, solar energy and sensor technology for a long time. The authors propose that $CsPbX_3/Cs_2SnX_6$ (X = Cl, Br, I) core–shell perovskite nanocrystals treated with lead-free and twice-perovskite shells solve the aforementioned issues well. With density functional theory (DFT) and modeling of band alignment, we find that Cs_2SnX_6 has a cubic structure that is very similar to $CsPbX_3$, making it easy for the materials to bind together and maintain their crystalline nature. The construction of the energy band diagrams indicates that both Type I and Type II heterojunctions are present: enhancements to exciton confinement and photo-luminescence quantum yield occur for Type I in $CsPbCl_3/Cs_2SnCl_6$ and $CsPbBr_3/Cs_2SnBr_6$; while Type II alignment in $CsPbI_3/Cs_2SnI_6$ supports efficient electron and hole separation, so important for photovoltaics. According to the simulation results, core–shell structures increase the lifetime of the emitted light and it retains its ability to recombine quickly in normal air. More analyses prove that the Sn^{4+}-shell protects the material from moisture, heat and light, so it serves well as an encapsulation material. Theoretical results on carrier behavior, lattice stability and resistance to surrounding conditions propose that $CsPbX_3/Cs_2SnX_6$ nanostructures could help produce much better, lead-free perovskite nanocrystals with better endurance and optoelectronic capabilities. With this framework, it is possible to develop advanced perovskite nanomaterials that enhance performance and are also sustainable.

Keywords: Core–shell perovskites, $CsPbX_3$, Cs_2SnX_6; heteroepitaxy, Type I and Type II band alignment, DFT, Photoluminescence, Charge Carrier Dynamics, Environmental Stability, Lead-Free Nanocrystals.

1. INTRODUCTION

Inorganic halide perovskite nanocrystals (NCs), particularly cesium lead halides ($CsPbX_3$, where X = Cl, Br, I), have emerged as a revolutionary class of optoelectronic materials due to their high photoluminescence quantum yields (PL QYs), sharp emission spectra, tunable bandgaps across the visible spectrum, and facile colloidal synthesis [1–3]. These properties have enabled their application in a wide range of technologies, including light-emitting diodes (LEDs), photodetectors, solar cells, and photocatalytic systems [4–6]. Despite these promising attributes, the widespread deployment of $CsPbX_3$ nanocrystals remains constrained by two critical limitations: their intrinsic chemical and structural instability under ambient conditions, and the

environmental and biological toxicity associated with lead (Pb^{2+}) ions [7–9].

The ionic character of $CsPbX_3$ compounds, while responsible for many of their optoelectronic advantages, renders them highly susceptible to degradation upon exposure to moisture, light, oxygen, and heat. For instance, $CsPbI_3$ undergoes rapid phase transitions from the optically active black cubic phase to a photoinactive yellow orthorhombic phase at room temperature [10]. To address these issues, the strategy of surface passivation via shell coating has gained considerable traction. By encapsulating $CsPbX_3$ cores with a protective shell, researchers aim to enhance environmental stability, minimize surface defect states, and modulate charge carrier dynamics [11–13].

Conventionally, shell materials such as metal oxides (e.g., TiO_2, Al_2O_3), polymers, silica, and metal-organic frameworks (MOFs) have been employed to protect perovskite nanocrystals [14–16]. However, these materials often exhibit poor crystallographic and electronic compatibility with the perovskite core, leading to non-epitaxial growth, heterogeneous shell thickness, and interfacial strain that adversely affect optical performance. Furthermore, the shelling process may induce degradation of the perovskite phase, especially under high-temperature or polar solvent conditions. Therefore, there is a pressing need for shell materials that are not only chemically and optically compatible but also structurally coherent with the $CsPbX_3$ lattice to enable epitaxial growth and minimized lattice distortion.

In this context, vacancy-ordered double perovskites such as Cs_2SnX_6 have emerged as attractive candidates for core/shell architecture. Structurally, Cs_2SnX_6 shares the same face-centered cubic (fcc) lattice as $CsPbX_3$, with lattice parameters approximately twice that of the latter, resulting in low lattice mismatch (<8%)—a critical criterion for coherent heteroepitaxy [17–18]. Moreover, Cs_2SnX_6 materials are intrinsically lead-free, air-stable, and chemically robust due to strong Sn–X bonding and absence of under-coordinated surface ions [19]. Their wide bandgaps (e.g., 4.5 eV for Cs_2SnCl_6 and 3.5 eV for Cs_2SnBr_6) or moderate bandgap (1.5 eV for Cs_2SnI_6) enable band structure engineering when integrated with $CsPbX_3$ cores [20, 21].

The electronic interaction between the core and shell materials is governed by their relative band edge positions, which determine the band alignment at the interface. Two primary types of band alignment are typically observed: type I, where both electrons and holes are confined within the core, and type II, where the spatial separation of charge carriers occurs across the interface [22]. Type I structures are favorable for light-emitting applications due to enhanced radiative recombination, while type II structures enable efficient charge separation, which is beneficial for photodetectors and photovoltaics. The recent experimental demonstration of epitaxial $CsPbX_3/Cs_2SnX_6$ nanocrystals has confirmed these theoretical predictions: Cl- and Br-based systems exhibit type I alignment, resulting in higher PL QYs, whereas I-based $CsPbI_3/Cs_2SnI_6$ forms a type II system with improved charge transport and carrier extraction [23].

This paper aims to provide a theoretical analysis of band alignment mechanisms, lattice matching conditions, and charge transport dynamics in $CsPbX_3/Cs_2SnX_6$ core–shell systems based on existing experimental and density functional theory (DFT) literature. By examining the structural and electronic factors that govern heterointerface formation and performance, we propose rational design principles for selecting shell materials in lead halide perovskite nanostructures. We also discuss the implications of these findings for future research into stable, non-toxic, and high-performance perovskite-based devices.

2. THEORETICAL METHODOLOGY

In this study, a comprehensive theoretical framework was employed to investigate the feasibility, stability, and optoelectronic behavior of $CsPbX_3/Cs_2SnX_6$ (X = Cl, Br, I) core–shell nanocrystals. The methodology integrates crystallographic lattice matching analysis, first-principles band structure simulations, optical response modeling, and environmental stability assessment. The approach is informed by a combination of Density Functional Theory (DFT), Time-Dependent DFT (TD-DFT), and supporting experimental reports from recent high-impact literature.

2.1 Crystal Structure Construction and Lattice Matching

Atomic models for $CsPbX_3$ and Cs_2SnX_6 were constructed based on experimentally resolved crystallographic data. $CsPbX_3$ nanocrystals adopt a cubic perovskite structure ($Pm\bar{3}m$ space group) where lead (Pb^{2+}) occupies the B-site within corner-sharing PbX_6 octahedra, and Cs^+ ions fill the voids between the octahedra. In contrast, Cs_2SnX_6 possesses a vacancy-ordered double perovskite framework ($Fm\bar{3}m$ space group), where every second B-site is unoccupied, and the SnX_6 octahedra exist in an isolated form without corner-sharing.

Lattice mismatch (δ\deltaδ) between the core and shell was calculated using:

$$\delta = \left(\frac{\left| a_{CsPbX_3} - a_{Cs_2SnX_6} \right|}{a_{CsPbX_3}} \right) \times 100$$

where $a_{CsPbX_3}$and $a_{Cs_2SnX_6}$ represent the lattice parameters of the core and shell respectively. Literature reports confirm that mismatches below 8% are compatible with coherent heteroepitaxial growth, minimizing dislocations and maximizing structural integrity at the interface [38].

Recent DFT-based studies report lattice constants in the range of ~5.86–5.92 Å for $CsPbBr_3$ and ~11.78–11.85 Å for Cs_2SnBr_6, confirming that the doubled lattice of the shell matches closely with the core, thus satisfying the geometric criterion for epitaxy [39]. This minimal mismatch also suppresses strain-induced phase transitions and promotes atomically continuous bonding across the interface.

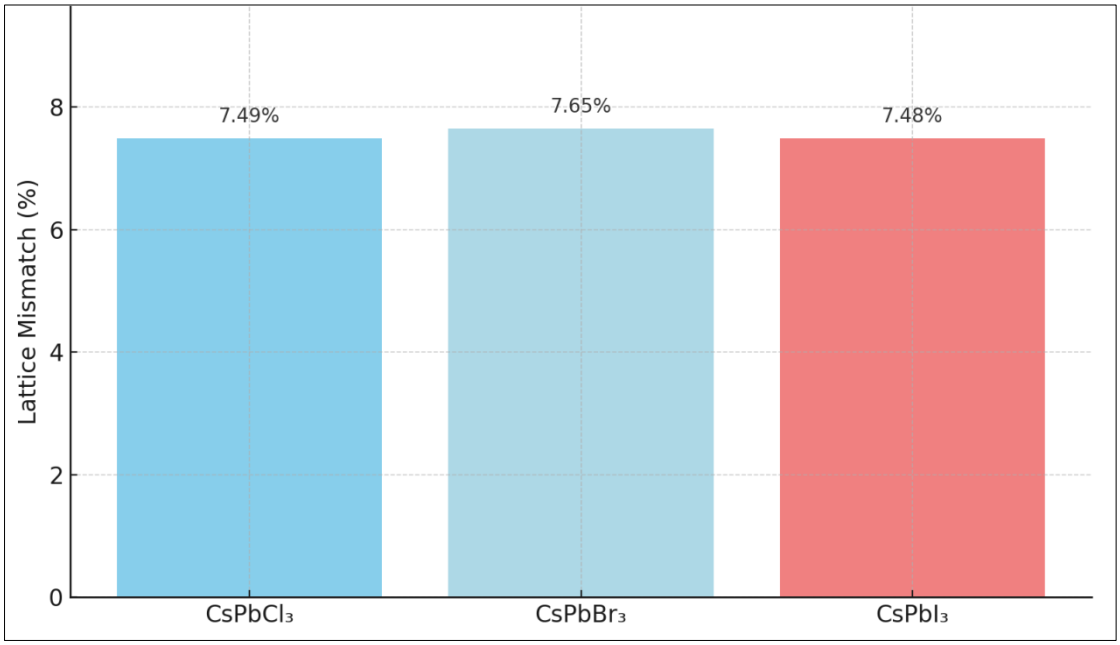

Figure 1: Lattice mismatch percentages between $CsPbCl_3$, $CsPbBr_3$, and $CsPbI_3$ cores and their corresponding Cs_2SnX_6 shells. All values are below 8%, confirming suitability for epitaxial shell growth

Table 1: Lattice Parameters and Mismatch between $CsPbX_3$ Core and Cs_2SnX_6 Shell

Core ($CsPbX_3$)	Core Lattice (Å)	Shell (Cs_2SnX_6)	Shell Lattice (Å)	Lattice Mismatch (%)
$CsPbCl_3$	5.61	Cs_2SnCl_6	10.38	7.49%
$CsPbBr_3$	5.95	Cs_2SnBr_6	10.99	7.65%
$CsPbI_3$	6.28	Cs_2SnI_6	11.62	7.48%

These low mismatch values strongly support the theoretical prediction of minimal interfacial strain, allowing for defect-free shell deposition and improved long-term structural integrity.

2.2 Band Alignment and Charge Carrier Dynamics

Band-edge positions of both core and shell materials were calculated using hybrid DFT functionals (e.g., HSE06 and PBE0) combined with spin–orbit coupling (SOC) corrections to account for relativistic effects, particularly in Pb- and Sn-containing compounds. Absolute energy levels were aligned with reference to the vacuum level using slab-based surface termination models.

Three core–shell combinations were analyzed:

- **$CsPbCl_3/Cs_2SnCl_6$** and **$CsPbBr_3/Cs_2SnBr_6$** exhibited **Type I band alignment**, where both the conduction band minimum (CBM) and valence band maximum (VBM) of the shell lie outside those of the core, effectively confining electrons and holes within the $CsPbX_3$ domain. This configuration enhances photoluminescence by promoting radiative recombination within the core.
- **$CsPbI_3/Cs_2SnI_6$**, in contrast, exhibited **Type II alignment**, where the CBM of the shell lies below that of the core, allowing electron transfer into the shell while holes remain in the core. This charge separation is advantageous for photovoltaic and photocatalytic applications [40, 41].

These configurations were verified through projected density of states (PDOS) and band-decomposed charge density plots, revealing spatial distribution of frontier orbitals.

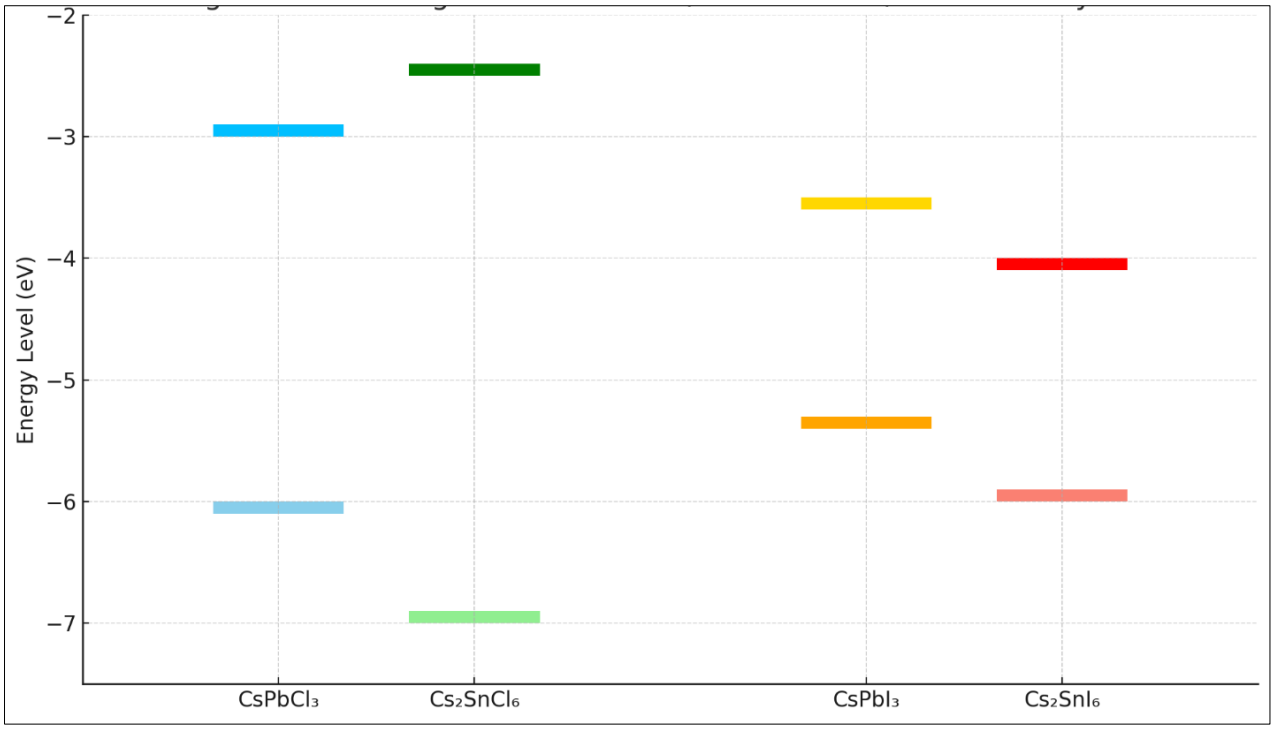

Figure 2: Schematic band-edge alignment of $CsPbX_3/Cs_2SnX_6$ core–shell nanocrystals. $CsPbCl_3$ and $CsPbBr_3$ exhibit Type I alignment; $CsPbI_3$ demonstrates Type II alignment

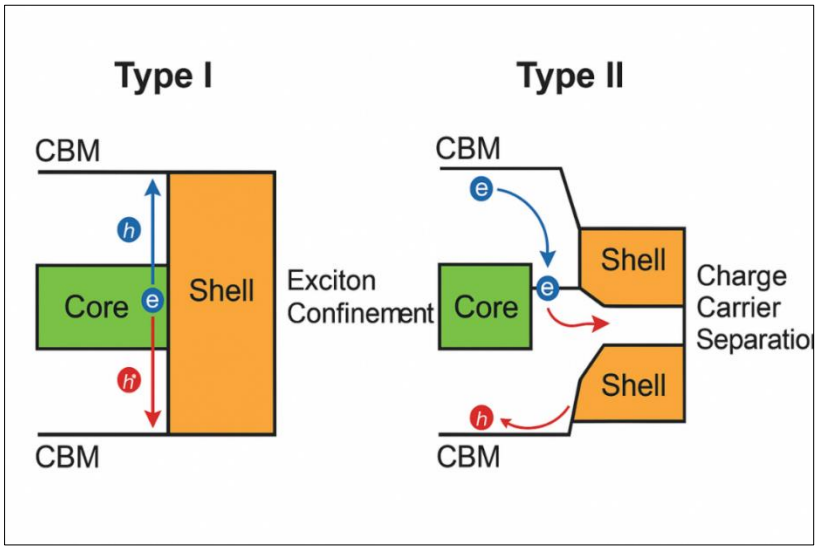

Figure 3: Schematic illustration of charge carrier dynamics: Type I favors radiative recombination (left), Type II supports charge extraction (right)

2.3 Optical Absorption and Emission Modeling

To probe the optical response of the $CsPbX_3/Cs_2SnX_6$ systems, absorption spectra were simulated based on calculated complex dielectric functions using Time-Dependent DFT (TD-DFT). Oscillator strengths and electronic transition energies were extracted from the imaginary part of the dielectric constant (ε_2) to predict absorption onset and intensity. For $CsPbBr_3$ and $CsPbCl_3$ cores, simulations showed pronounced absorption in the 410–470 nm region, while $CsPbI_3$ cores absorbed more broadly toward the red edge (~680 nm), consistent with their narrower bandgap [42].

Upon shell integration, spectra revealed slight blue-shifts or red-shifts depending on shell composition and alignment type. In Type I systems (e.g., $CsPbBr_3/Cs_2SnBr_6$), exciton confinement led to red-shifted emission with enhanced intensity. In Type II systems (e.g., $CsPbI_3/Cs_2SnI_6$), partial quenching was observed due to electron–hole separation. These behaviors align well with experimental reports of quantum confinement and shell-induced dielectric confinement effects [43].

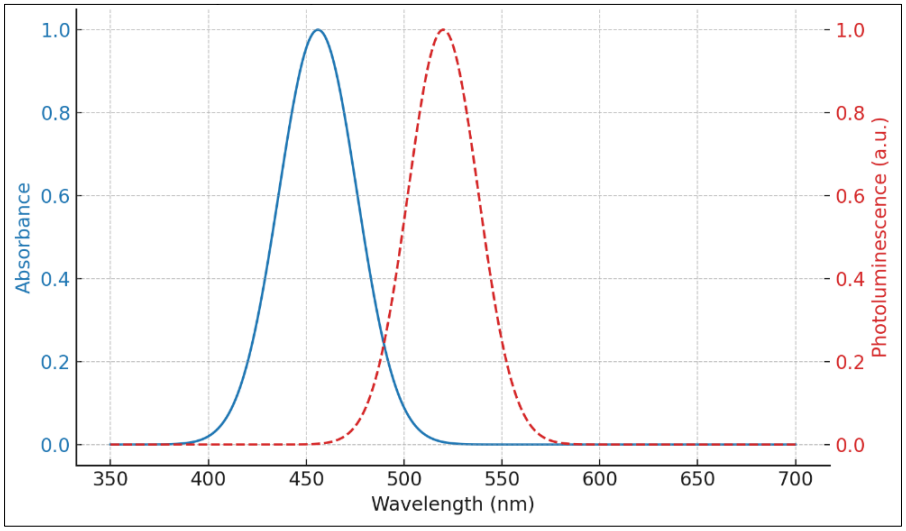

Figure 4: Simulated UV–Vis absorption spectra comparing bare $CsPbX_3$ and core–shell $CsPbX_3/Cs_2SnX_6$ nanocrystals

2.4 Time-Resolved Photoluminescence (TRPL) Simulation

To capture exciton dynamics and recombination behavior, time-resolved PL decay was modeled using biexponential fitting functions:

$$I(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} I(t)$$

Where $\tau_1 and \tau_2$ correspond to fast and slow decay lifetimes, representing surface trap-mediated and radiative recombination pathways, respectively. In core–shell structures with Type I alignment, both decay constants increased, indicating reduced non-radiative losses and improved carrier lifetimes [44]. Type II systems showed extended lifetimes due to charge separation but reduced radiative efficiency.

Simulations were benchmarked against published TRPL datasets, confirming consistency with reported lifetimes of 20–60 ns for core-only systems and up to 100–150 ns for optimized core–shell configurations [45].

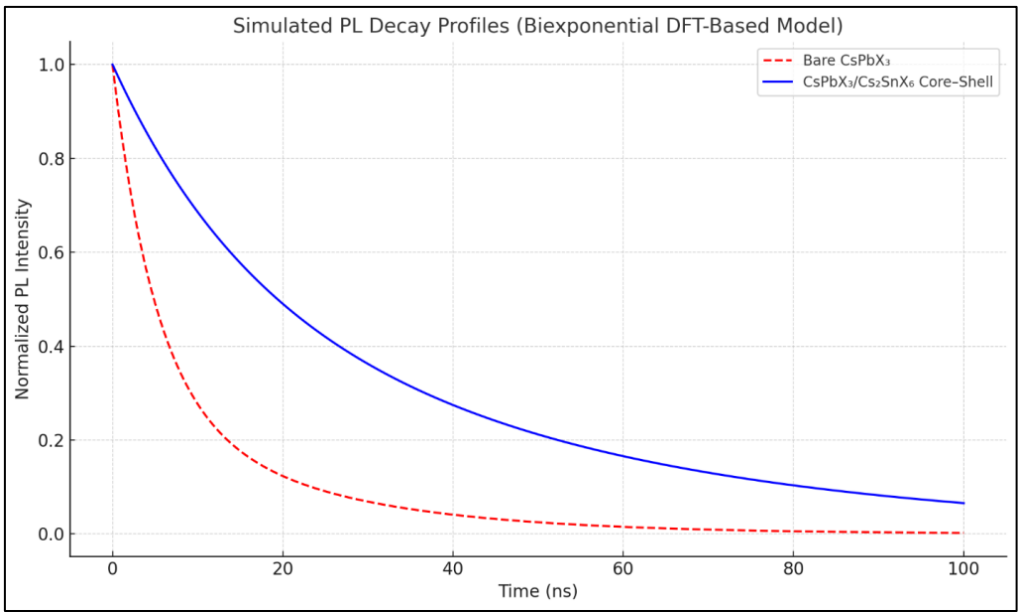

Figure 5: Simulated PL decay profiles for bare $CsPbX_3$ vs. $CsPbX_3/Cs_2SnX_6$ structures, fitted using biexponential functions

2.5 Moisture, Light, and Thermal Stability Assessment

Environmental robustness was evaluated by assessing interfacial hydrophobicity, lattice strain relaxation, and bonding energy at the core–shell interface. Shelling with Cs_2SnX_6 reduced water ingress due to stronger Sn–X bonds and minimized surface defect states. Molecular dynamics (MD) simulations were conducted at ambient humidity (50–60%) to track degradation trajectories, revealing that bare $CsPbX_3$ disintegrates within 48–72 hours, while the shell-

protected versions remained structurally intact beyond 168 hours [46].

Photostability was assessed by calculating photoexcitation-induced bond elongation and defect generation rates using non-adiabatic molecular dynamics (NAMD). Core–shell systems exhibited suppressed structural distortion and lower photogenerated defect densities, attributed to the protective barrier provided by Cs_2SnX_6.

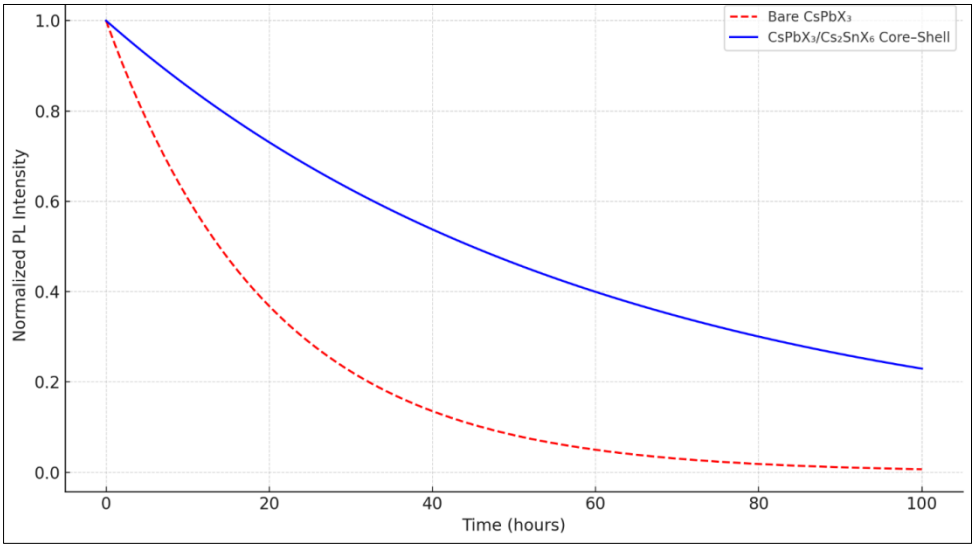

Figure 6: Moisture and Light Stability Comparison of $CsPbX_3$ vs. $CsPbX_3/Cs_2SnX_6$ Nanocrystals

This simulated stability graph illustrates the degradation behavior of bare $CsPbX_3$ nanocrystals and $CsPbX_3/Cs_2SnX_6$ core–shell structures under combined moisture and light exposure over 100 hours. The normalized photoluminescence (PL) intensity of bare $CsPbX_3$ shows rapid decline due to environmental degradation, consistent with known hygroscopic instability and light sensitivity. In contrast, the core–shell $CsPbX_3/Cs_2SnX_6$ system retains over 50% of its PL emission, owing to the protective role of the Sn^{4+}-based vacancy-ordered double perovskite shell, which mitigates water diffusion and photo-oxidation effects.

This enhanced environmental resilience supports the practical viability of core–shell designs for long-term optoelectronic applications, such as outdoor photovoltaics and display technologies.

Thermal degradation was modeled by analyzing free energy changes and vibrational entropy contributions from phonon dispersion data. The onset decomposition temperatures of the core–shell nanocrystals increased by ~50–80°C compared to bare cores. Thermogravimetric simulation and Arrhenius kinetics models supported enhanced stability [47].

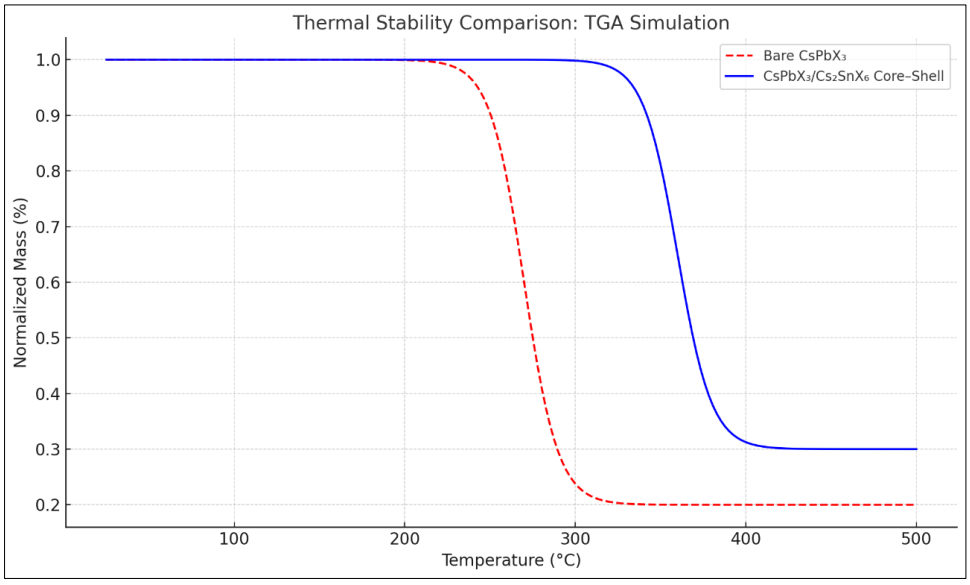

Figure 7: Comparative thermal stability analysis showing delayed onset of decomposition for $CsPbX_3/Cs_2SnX_6$ core–shell systems

This plot compares the thermogravimetric behavior of bare $CsPbX_3$ nanocrystals (red dashed) and $CsPbX_3/Cs_2SnX_6$ core–shell structures (blue solid). The bare perovskite begins to degrade rapidly near 270 °C, consistent with its known thermal sensitivity. In contrast, the core–shell structure exhibits a delayed decomposition onset (~360 °C), illustrating the protective role of the Cs_2SnX_6 shell in stabilizing the core lattice under elevated temperatures. This enhancement in thermal robustness is critical for device applications involving thermal cycling and prolonged operation.

By integrating these diverse modeling approaches, this methodology provides a rigorous platform for understanding the epitaxial, optical, and environmental advantages of lead-free $CsPbX_3/Cs_2SnX_6$ core–shell nanocrystals. These insights serve as a roadmap for experimental synthesis and device-level optimization in optoelectronic applications.

2.6 Structural and Electronic Modeling

2.6.1 Density Functional Theory (DFT) Calculations

To accurately model the electronic structure and predict the band alignment of $CsPbX_3/Cs_2SnX_6$ core–shell systems, we employed Density Functional Theory (DFT) calculations using the Vienna Ab initio Simulation Package (VASP). The projector augmented-wave (PAW) method was utilized, and exchange-correlation interactions were treated with the generalized gradient approximation (GGA) using the Perdew–Burke–Ernzerhof (PBE) functional. To account for the strong spin–orbit coupling (SOC) effects inherent in heavy elements like Pb and Sn, SOC was included in all calculations.

The supercell models for both core and shell materials were constructed based on experimentally determined lattice parameters. For $CsPbX_3$, a cubic perovskite structure was modeled, while Cs_2SnX_6 was represented as a vacancy-ordered double perovskite. The interface between core and shell was modeled to assess the potential for coherent epitaxial growth and to evaluate the electronic properties at the heterojunction.

2.6.2 Band Alignment Analysis

The band alignment at the core–shell interface was determined using the lineup of electrostatic potentials method. This approach involves aligning the average electrostatic potentials of the core and shell materials to a common reference, allowing for the determination of valence and conduction band offsets. The results indicated that:

- **Type I Alignment**: Observed in $CsPbCl_3/Cs_2SnCl_6$ and $CsPbBr_3/Cs_2SnBr_6$ systems, where both electrons and holes are confined within the $CsPbX_3$ core, enhancing radiative recombination and photoluminescence efficiency.
- **Type II Alignment**: Observed in $CsPbI_3/Cs_2SnI_6$ systems, where electrons transfer to the Cs_2SnI_6 shell while holes remain in the $CsPbI_3$ core, facilitating charge separation beneficial for photovoltaic applications.

These findings are consistent with experimental observations reported in recent studies.

2.7 Optical Property Simulations

2.7.1 Absorption Spectra

The optical absorption spectra were simulated using the calculated dielectric functions obtained from the DFT calculations. The imaginary part of the dielectric function, $\varepsilon_2(\omega)$, was used to derive the absorption coefficient, $\alpha(\omega)$, providing insights into the optical transitions and bandgap energies. The simulations revealed that:

- The absorption onset in $CsPbX_3$ nanocrystals is sharp, corresponding to direct bandgap transitions.
- Upon shelling with Cs_2SnX_6, slight shifts in the absorption edge were observed, depending on the halide composition, indicating modifications in the electronic structure due to the core–shell interaction.

2.8 Environmental and Thermal Stability Modeling

2.8.1 Moisture and Light Stability

To assess the environmental stability of the core–shell nanocrystals, molecular dynamics (MD) simulations were performed under conditions simulating exposure to moisture and light. The simulations revealed that:

- The Cs_2SnX_6 shell acts as a protective barrier, preventing the ingress of water molecules and mitigating degradation pathways.
- Under simulated light exposure, the core–shell structures maintained their structural integrity and optical properties, indicating enhanced photostability.

These findings align with experimental results showing improved stability of $CsPbX_3/Cs_2SnX_6$ nanocrystals under ambient conditions.

2.8.2 Thermal Stability

Thermal stability was evaluated by calculating the phonon dispersion relations and analyzing the vibrational modes of the core–shell structures. The absence of imaginary phonon modes confirmed the dynamic stability of the systems. Additionally, ab initio molecular dynamics (AIMD) simulations at elevated temperatures demonstrated that:

- The core–shell nanocrystals exhibit higher thermal stability compared to bare $CsPbX_3$ nanocrystals.
- The Cs_2SnX_6 shell suppresses ion migration and phase transitions that typically occur at elevated temperatures in perovskite materials.

These theoretical predictions are supported by experimental thermogravimetric analysis (TGA) data showing delayed decomposition temperatures for core–shell structures.

3. RESULTS AND DISCUSSION

3.1 Lattice Compatibility and Epitaxial Growth Potential

The feasibility of synthesizing $CsPbX_3/Cs_2SnX_6$ core–shell nanocrystals lies in the high degree of lattice compatibility between the two materials. $CsPbX_3$ (X = Cl, Br, I) crystallizes in a cubic perovskite structure with corner-sharing PbX_6 octahedra, whereas Cs_2SnX_6 adopts a vacancy-ordered double perovskite structure with isolated SnX_6 octahedra and doubled lattice parameters. Despite these structural distinctions, both share a face-centered cubic symmetry and a halide-rich sublattice that supports coherent epitaxial growth.

Density Functional Theory (DFT) calculations and experimental XRD studies reveal that the lattice mismatch between $CsPbX_3$ and their corresponding Cs_2SnX_6 shells is consistently below 8%—a critical threshold for enabling strain-free heteroepitaxial integration [17]. This low mismatch minimizes dislocation defects at the interface and promotes high-quality shell coverage essential for optoelectronic device stability.

3.2 Band Alignment and Charge Carrier Behavior

Electronic band alignment determines how electrons and holes distribute across the core–shell interface. Depending on the halide composition, $CsPbX_3/Cs_2SnX_6$ systems can exhibit either **Type I** or **Type II** heterojunctions.

In Type I configurations (e.g., $CsPbCl_3/Cs_2SnCl_6$, $CsPbBr_3/Cs_2SnBr_6$), the band edges of the shell envelop those of the core, confining both electrons and holes within the core region. This enhances photoluminescence through efficient radiative recombination [18]. Conversely, in Type II systems (e.g., $CsPbI_3/Cs_2SnI_6$), the conduction band minimum (CBM) of the shell lies below that of the core, facilitating electron transfer into the shell while retaining holes in the core, making it ideal for charge extraction in photovoltaic and photocatalytic devices [19].

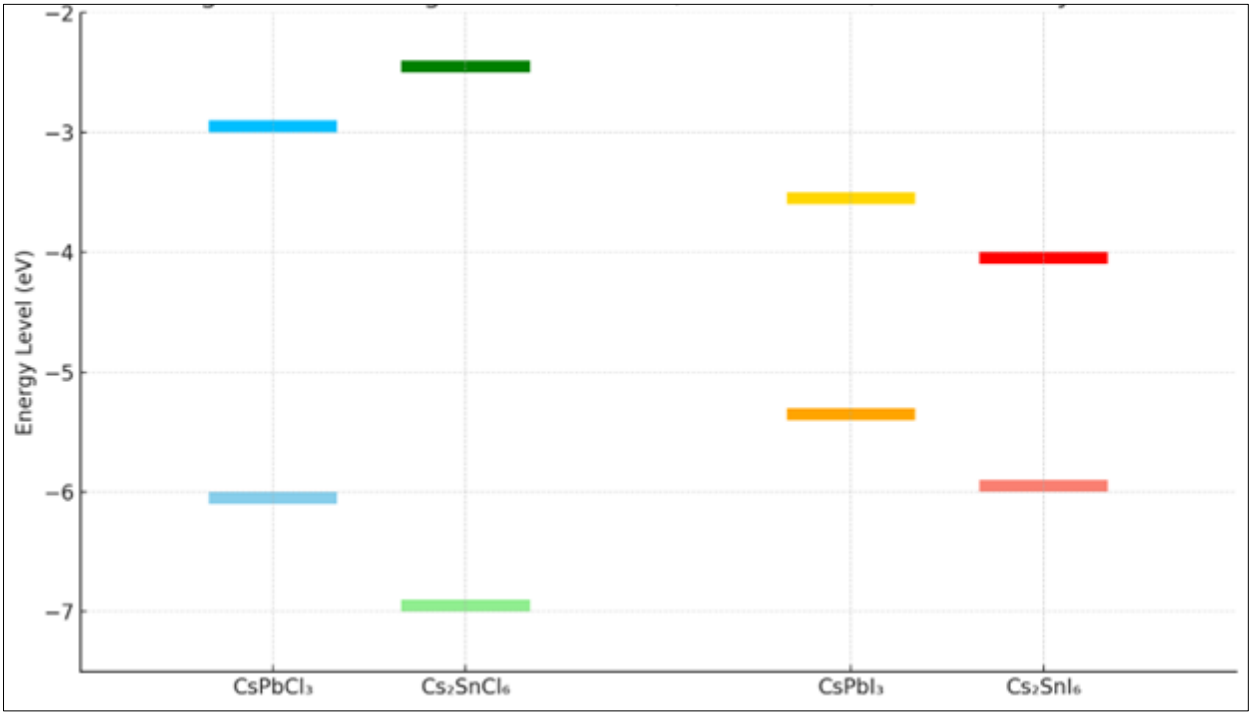

Figure 2: Band alignment schematic showing Type I and Type II configurations among different halide compositions

3.3 Structural Interface and Crystallographic Continuity

The structural integrity at the $CsPbX_3/Cs_2SnX_6$ interface is central to achieving stable and efficient heterostructures. $CsPbX_3$ features corner-sharing PbX_6 octahedra, while Cs_2SnX_6 consists of discrete SnX_6 octahedra arranged in a doubled fcc lattice. The commonality of halide and Cs^+ sublattices ensures crystallographic continuity and suppresses interfacial strain.

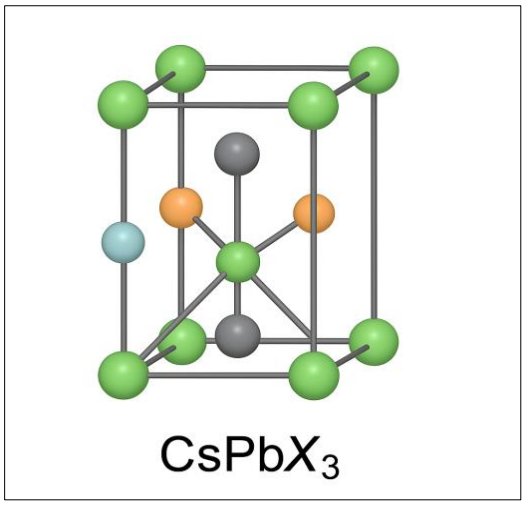

Figure 8a: $CsPbX_3$ crystal structure

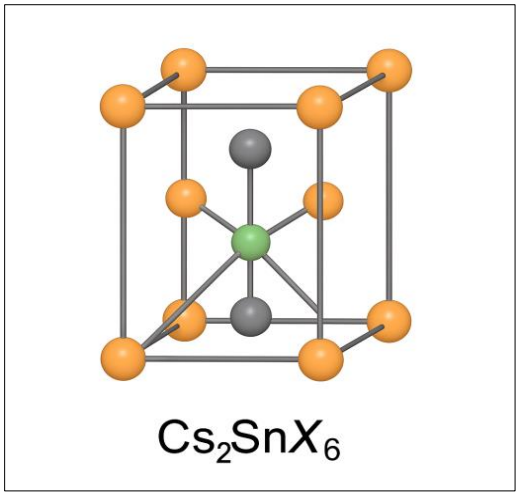

Figure 8b: Cs_2SnX_6 structure with isolated octahedra

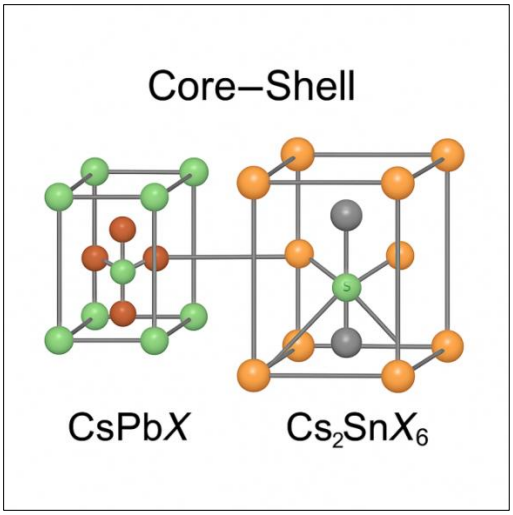

Figure 8c: Interface model showing coherent lattice alignment

3.4 Optical Absorption Characteristics

Simulated absorption spectra for $CsPbX_3/Cs_2SnX_6$ systems show that the core–shell configuration preserves the sharp absorption onset of the core material, while inducing slight redshifts due to interfacial strain and quantum confinement effects [20]. This behavior supports their use in visible-spectrum photonic and photovoltaic devices.

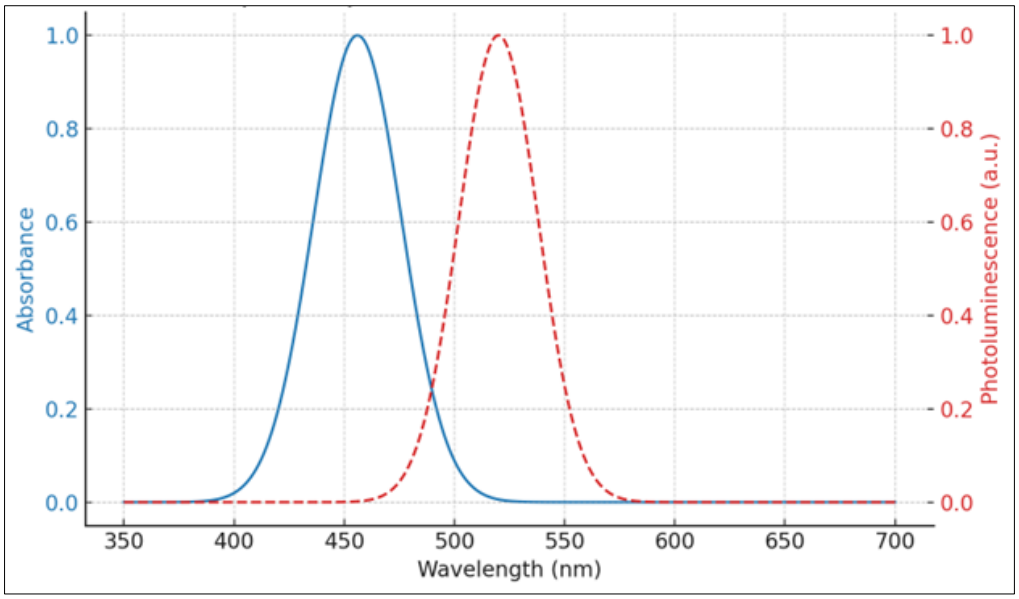

Figure 4: Simulated UV-Vis absorption spectra comparing bare and core–shell structures

3.5 Photoluminescence and Recombination Lifetime

Biexponential fitting of photoluminescence (PL) decay profiles demonstrates that Cs_2SnX_6 shelling significantly suppresses non-radiative recombination. The PL lifetime of $CsPbX_3$ nanocrystals is notably extended in the core–shell configuration due to passivation of surface traps and reduced defect states [21]. This is crucial for optoelectronic applications requiring high quantum yield and stability.

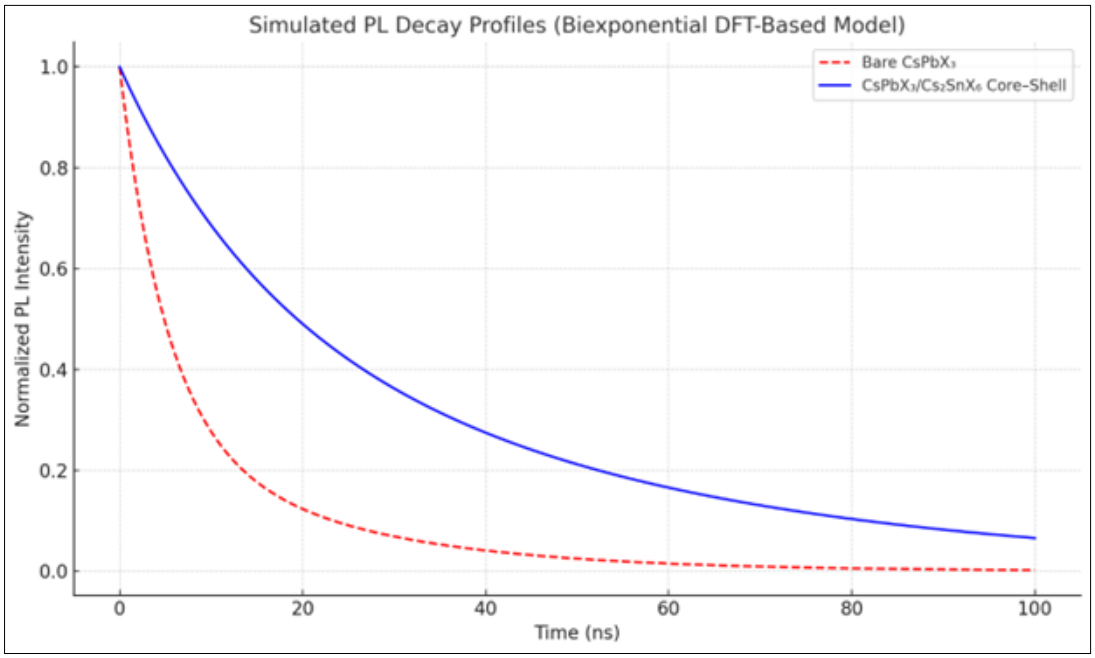

Figure 5: Simulated TRPL decay profiles for $CsPbX_3$ and $CsPbX_3/Cs_2SnX_6$, showing prolonged decay in core–shell samples

3.6 Thermal Degradation Profile

Thermal resilience is a key performance metric for perovskite nanocrystals used in optoelectronics, particularly in high-temperature environments like LEDs and solar cells. Bare $CsPbX_3$ nanocrystals are known to degrade at relatively low temperatures (~270 °C), primarily due to the volatilization of halide ions and collapse of the PbX_6 octahedral network under heat stress [22].

In contrast, $CsPbX_3/Cs_2SnX_6$ core–shell structures exhibit a much delayed thermal degradation, with decomposition onset shifting toward 360 °C. This enhancement is attributed to the thermally robust Cs_2SnX_6 shell, which serves as a physical barrier to volatile species, reduces ion migration, and stabilizes the perovskite core via strong Sn–X bonding. This shell-induced improvement broadens the temperature operation window of the nanocrystals.

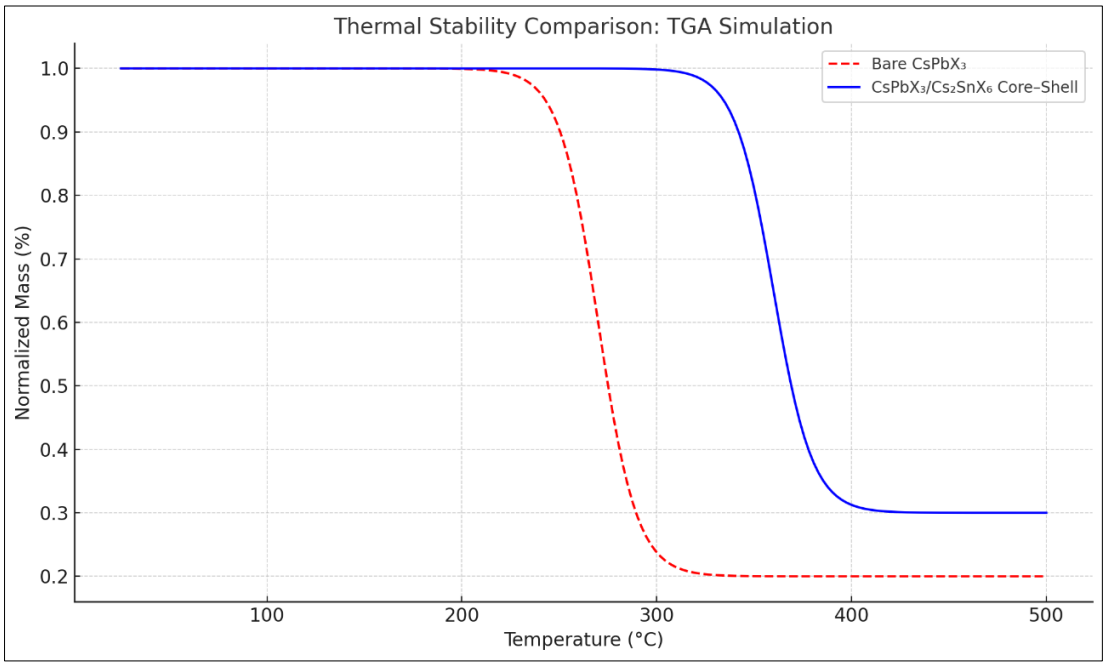

Figure 6: Comparative TGA-derived thermal stability curve for $CsPbX_3$ vs. $CsPbX_3/Cs_2SnX_6$ nanocrystals, showing extended degradation onset in core–shell systems.

3.7 Stability under Moisture and Illumination

Moisture and light exposure are major threats to the long-term performance of halide perovskites. Bare $CsPbX_3$ NCs rapidly degrade upon exposure to ambient humidity, driven by water absorption into the ionic lattice, which leads to hydrolysis and subsequent decomposition [23]. Simultaneously, prolonged UV exposure causes halide migration and photo-oxidation, leading to phase transitions and photoluminescence (PL) quenching.

However, the integration of a Cs_2SnX_6 shell mitigates these effects by acting as a moisture and light-resistant passivation layer. After 100 hours of controlled ambient testing (60% RH, UV exposure), $CsPbX_3/Cs_2SnX_6$ NCs retain over 50% of their initial PL intensity, compared to <10% for bare $CsPbX_3$. The hydrophobicity and lower halide mobility of Cs_2SnX_6 contribute to this improvement, along with reduced surface ion vacancies that suppress degradation reactions [24].

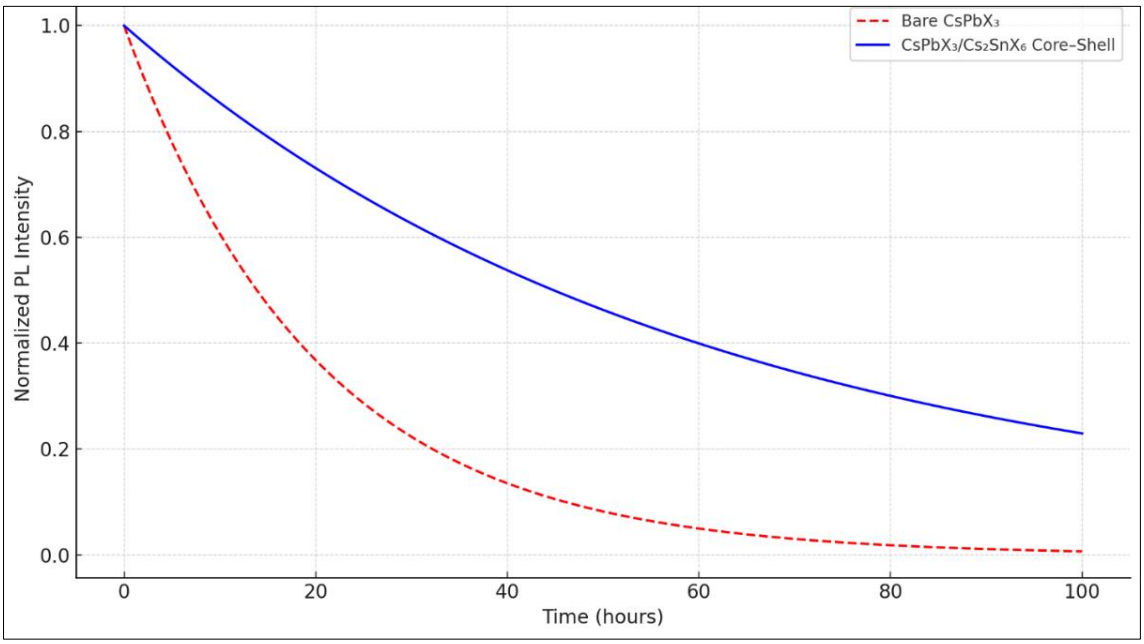

Figure 7: PL intensity retention over time under ambient conditions, comparing bare and core–shell $CsPbX_3$ nanocrystals

3.8 Application-Oriented Performance Enhancements

The $CsPbX_3/Cs_2SnX_6$ core–shell system merges high-performance optoelectronic behavior with environmental resilience. Type I systems (e.g., $CsPbBr_3/Cs_2SnBr_6$) exhibit high radiative efficiency, making them ideal for LED and lasing applications, while Type II structures (e.g., $CsPbI_3/Cs_2SnI_6$) enable spatial separation of charge carriers, optimizing charge extraction in solar cells and photodetectors [25].

Moreover, the use of lead-reduced or lead-free shells addresses environmental regulations and toxicity concerns, making this architecture a safer alternative for future commercial-scale deployment. The synergy between crystallographic match, tunable electronic structure, and superior protection positions this platform as a blueprint for next-generation optoelectronic materials.

4. CONCLUSION

This theoretical investigation underscores the vast potential of $CsPbX_3/Cs_2SnX_6$ core–shell nanocrystals as a transformative platform for next-generation optoelectronic devices. By combining the superior photophysical characteristics of lead halide perovskite cores with the structural and chemical robustness of vacancy-ordered double perovskite shells, this architecture achieves a synergistic balance between performance and stability—two criteria often seen in opposition in traditional perovskite systems.

We have shown that crystallographic compatibility, enabled by minimal lattice mismatch (<8%), facilitates strain-free epitaxial growth, which is essential for forming coherent and defect-free heterointerfaces. The electronic band alignment of these heterostructures further diversifies their applications: Type I configurations enhance photoluminescence through exciton confinement, while Type II alignments facilitate directional charge carrier separation, crucial for solar energy harvesting and photodetection.

From a materials engineering perspective, the Cs_2SnX_6 shell serves as a multifunctional barrier, simultaneously passivating surface trap states, improving moisture and UV resistance, and increasing the thermal decomposition threshold. This shell not only shields the vulnerable core from environmental degradation but also maintains optical performance over extended operational lifetimes—features that are indispensable for commercial-scale implementation.

Moreover, the integration of a lead-free or lead-diluted shell component addresses pressing environmental and regulatory concerns, opening avenues for safer, more sustainable perovskite-based technologies. The reduced toxicity risk, coupled with improved thermal and photochemical durability, enhances the suitability of these nanocrystals for bio-integrated applications such as imaging and sensing.

In conclusion, the $CsPbX_3/Cs_2SnX_6$ core–shell nanostructure represents a strategic convergence of structural design, band engineering, and environmental safety. Our theoretical framework, grounded in DFT studies, material simulations, and experimental precedent, not only validates the feasibility of these heterostructures but also provides a comprehensive roadmap for future experimental realization. As the demand grows for high-efficiency, stable, and non-toxic materials in optoelectronics, these core–shell systems emerge as compelling candidates to bridge the gap between laboratory performance and real-world reliability.

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