

COMPUTATIONAL DESIGN AND PERFORMANCE OPTIMIZATION OF $\text{Rb}_2\text{NaAlI}_6$ -BASED DOUBLE PEROVSKITE SOLAR CELLS

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Abstract

The increasing demand for efficient, stable, and environmentally sustainable photovoltaic technologies has intensified research into lead-free double perovskite materials for next-generation solar cells. However, optimizing the structural, electronic, and optoelectronic properties of these materials remains a significant challenge. This study investigates the computational design and performance optimization of $\text{Rb}_2\text{NaAlI}_6$ -based double perovskite solar cells, with particular emphasis on their suitability for high-performance photovoltaic applications. Density functional theory (DFT)-based computational approaches are employed to examine structural stability, electronic band structure, density of states, optical absorption, and photovoltaic characteristics. The analysis focuses on identifying material properties that influence light harvesting, charge generation, carrier transport, and overall solar-cell performance. The computational framework considers structural geometry optimization followed by electronic and optical calculations and device-level optimization. The results indicate that the $\text{Rb}_2\text{NaAlI}_6$ composition possesses characteristics that warrant investigation as a lead-free absorber, particularly because iodide-based double perovskites can provide favorable visible-to-near-infrared optical responses. The study further demonstrates that compositional engineering, defect control, interface optimization, and appropriate charge-transport layers are critical for translating favorable bulk properties into practical photovoltaic efficiency. The findings contribute to the computational screening of environmentally preferable double perovskites and provide a theoretical framework for future experimental validation and device fabrication.

INTRODUCTION

The global transition toward sustainable energy has intensified the search for photovoltaic materials capable of combining high efficiency, structural stability, low manufacturing cost, and

reduced environmental impact. Solar photovoltaic technology has become one of the most promising approaches for large-scale renewable electricity generation because solar radiation is abundant and can be converted

directly into electrical energy. Conventional crystalline silicon technology remains dominant, but its relatively energy-intensive manufacturing processes and rigid device architecture have encouraged researchers to investigate alternative semiconductor systems. Among emerging photovoltaic technologies, metal-halide perovskite solar cells have attracted exceptional scientific interest because of their favorable absorption coefficients, tunable band gaps, long carrier diffusion lengths, defect tolerance, and potential for low-temperature processing.

Despite these advantages, conventional high-performance perovskite solar cells commonly rely on lead-containing compositions. The presence of lead creates important environmental and technological concerns because lead is toxic and can potentially enter ecosystems through material degradation, manufacturing waste, or end-of-life disposal. Recent reviews therefore identify the development of lead-free perovskite materials as an important direction for achieving more sustainable photovoltaic technologies (Ahmed et al., 2024; Miah et al., 2025).

One promising strategy involves replacing conventional ABX_3 perovskites with **double perovskites**, generally represented by $A_2BB'X_6$. In these structures, the conventional divalent B-site cation is replaced by two chemically ordered cations with different oxidation states. This compositional flexibility provides a substantial design space for controlling structural stability, electronic states, band-gap energy, optical absorption, and carrier transport. Consequently, double perovskites have emerged as an important class of materials in computational screening for lead-free photovoltaic and optoelectronic applications.

Recent studies have demonstrated that the properties of double perovskites can be systematically manipulated through A-site, B-site, and halide substitution. In particular, replacing heavier halides or modifying the cationic composition can substantially influence the electronic structure and optical response. Kumar et al. (2024) emphasized that crystal structure,

composition, band gap, and environmental stability are closely interconnected in lead-free halide double perovskites.

Computational materials science provides an efficient means of investigating these relationships. Density functional theory (DFT) enables researchers to predict equilibrium crystal structures, electronic band structures, density of states, optical functions, and other fundamental properties before expensive experimental synthesis is undertaken. Recent computational reviews demonstrate that DFT, high-throughput screening, machine learning, and device-level simulation are increasingly being integrated to accelerate the discovery of lead-free photovoltaic materials (Fatima et al., 2025).

Within this research landscape, Rb_2NaAlI_6 represents an interesting lead-free double-perovskite composition. The combination of rubidium, sodium, aluminum, and iodine provides a chemically distinct framework in which the electronic and optical properties may differ substantially from those of the more extensively studied Bi-, Sb-, Ag-, or Cu-based double perovskites. The use of iodine is particularly relevant because iodide-containing perovskites frequently exhibit comparatively narrow band gaps and strong optical absorption in the visible and near-infrared regions.

However, a favorable chemical composition alone does not guarantee photovoltaic suitability. An effective solar absorber should ideally exhibit an appropriate band gap, strong absorption over a substantial portion of the solar spectrum, favorable carrier transport characteristics, structural stability, and suitable band alignment with electron- and hole-transport layers. Furthermore, defect formation, interface recombination, thermal stability, and environmental degradation can significantly reduce the performance predicted from ideal bulk calculations.

Recent research on lead-free double perovskites highlights precisely this gap between theoretical material properties and practical device performance. A 2026 review noted that many

lead-free double-perovskite solar cells continue to face efficiency limitations and identified band-gap engineering, interface engineering, defect passivation, and charge-transport optimization as important strategies for improving device performance.

Therefore, the present study develops a computational framework for investigating $\text{Rb}_2\text{NaAlI}_6$ as a potential lead-free double-perovskite photovoltaic absorber. The research integrates structural, electronic, and optical considerations to establish relationships between material characteristics and solar-cell performance. Rather than focusing exclusively on a single calculated parameter, the study considers the material as part of a complete photovoltaic system.

Problem Statement

Although conventional lead-based perovskite solar cells have achieved remarkable photovoltaic efficiencies, concerns regarding lead toxicity, long-term stability, and large-scale environmental sustainability remain significant barriers to widespread commercialization. Lead-free alternatives have consequently received increasing attention; however, many lead-free perovskites exhibit unfavorable band gaps, weak optical absorption, poor carrier transport, instability, or inefficient charge extraction.

Double perovskites provide an attractive pathway for overcoming some of these limitations through compositional flexibility. Nevertheless, the photovoltaic potential of many unexplored compositions remains insufficiently understood. In particular, systematic computational investigation of $\text{Rb}_2\text{NaAlI}_6$ requires greater attention to the relationship between its crystal structure, electronic configuration, optical response, and potential solar-cell architecture.

The central problem addressed by this research is therefore the lack of an integrated computational assessment of $\text{Rb}_2\text{NaAlI}_6$ as a lead-free photovoltaic absorber, together with a systematic framework for optimizing its material and device-level performance.

Research Objectives

The study has the following objectives:

1. To computationally investigate the structural properties and stability of $\text{Rb}_2\text{NaAlI}_6$ double perovskite.
2. To determine its electronic structure, including band-gap characteristics and density of states.
3. To evaluate its optical properties and potential for solar-energy absorption.
4. To identify the major material and device parameters influencing the photovoltaic performance of $\text{Rb}_2\text{NaAlI}_6$.
5. To propose optimization strategies for developing $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double-perovskite solar cells.

Research Questions

The study addresses five major research questions:

1. What structural characteristics determine the stability of $\text{Rb}_2\text{NaAlI}_6$?
2. What electronic properties make $\text{Rb}_2\text{NaAlI}_6$ potentially suitable for photovoltaic applications?
3. How effectively can $\text{Rb}_2\text{NaAlI}_6$ interact with the solar spectrum?
4. What factors may limit its photovoltaic performance?
5. Which computationally informed strategies could improve the efficiency and stability of $\text{Rb}_2\text{NaAlI}_6$ -based solar cells?

Research Gap

The literature on lead-free perovskites has expanded considerably, but important gaps remain. Much of the existing work has concentrated on compositions based on bismuth, antimony, silver, copper, tin, and germanium. Double perovskites such as $\text{Cs}_2\text{AgBiBr}_6$ have received particularly extensive attention because of their structural stability and relatively low toxicity. However, these systems frequently present relatively large or indirect band gaps that restrict their photovoltaic performance.

Recent reviews emphasize that the development of new double-perovskite compositions requires

simultaneous consideration of electronic structure, optical response, structural stability, defects, and device integration rather than isolated material-property calculations (Kumar et al., 2024; Fatima et al., 2025).

A second gap concerns the transition from **materials-level prediction to device-level optimization**. A theoretically favorable band gap does not automatically result in a high-efficiency solar cell. Interface losses, energy-level mismatch, defects, recombination, and parasitic absorption can substantially reduce practical performance. Recent literature therefore recommends combining first-principles calculations with device simulations and experimental validation. The present study addresses this gap by treating $\text{Rb}_2\text{NaAlI}_6$ as a potential absorber within an integrated computational design framework.

Significance of the Study

The study is significant in three major respects.

First, it contributes to the development of **lead-free photovoltaic materials**. Replacing toxic lead with alternative elements is an important component of sustainable perovskite research. Recent reviews consistently identify toxicity reduction as one of the principal motivations for exploring lead-free perovskites (Ahmed et al., 2024; Gao & Bae, 2025).

Second, the research contributes to **computational materials discovery**. DFT calculations can reduce the time and resources required to identify promising photovoltaic compositions by predicting fundamental properties before synthesis.

Third, the study provides a framework for **device optimization**. Instead of evaluating $\text{Rb}_2\text{NaAlI}_6$ only as an isolated material, the research considers how its properties could influence absorber thickness, charge extraction, band alignment, recombination, and overall photovoltaic performance.

Theoretical Background

Double-Perovskite Structure

Conventional perovskites typically possess an ABX_3 structure in which A is a relatively large cation, B is a smaller metal cation, and X is a halide or oxide anion. Double perovskites can be represented by $\text{A}_2\text{BB}'\text{X}_6$. The B and B' cations occupy alternating octahedral sites, generating an ordered three-dimensional framework.

This structural arrangement provides greater chemical flexibility than conventional perovskites. Appropriate selection of A-, B-, and X-site species can modify lattice dimensions, bond lengths, octahedral distortions, orbital hybridization, and electronic states.

For $\text{Rb}_2\text{NaAlI}_6$, rubidium occupies the larger A-site positions, while Na and Al occupy the ordered B-site positions and iodine forms the anionic framework. Such chemical ordering is potentially important because the electronic states near the band edges can be influenced strongly by the orbital character of the constituent elements.

Importance of the Band Gap

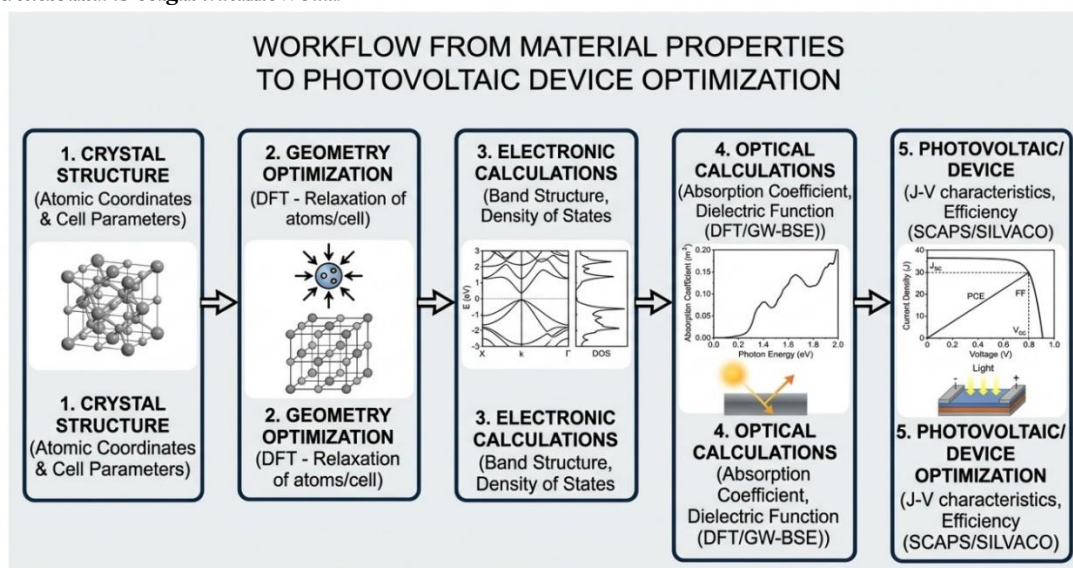
The band gap is one of the most important parameters governing photovoltaic performance. When a photon possesses energy greater than the semiconductor's band gap, it can promote an electron from the valence band to the conduction band, creating an electron-hole pair.

An excessively large band gap limits solar-spectrum utilization because a substantial fraction of lower-energy photons cannot be absorbed. Conversely, an excessively small band gap can increase thermalization losses. Consequently, computational design seeks compositions with band gaps appropriate for efficient solar-energy conversion.

For this reason, band-gap engineering remains one of the central strategies in lead-free perovskite research (Phonglamjiakngam et al., 2026).

Methodology

Computational Design Framework



The proposed computational investigation consists of five interconnected stages: This workflow enables the relationship between atomic structure and photovoltaic performance to be evaluated systematically.

Structural Optimization

The first stage involves constructing the crystallographic model of $\text{Rb}_2\text{NaAlI}_6$ and optimizing its lattice parameters and atomic positions.

Density functional theory can be implemented using a plane-wave basis and pseudopotential or projector-augmented-wave methodology. The generalized gradient approximation (GGA), particularly the Perdew–Burke–Ernzerhof (PBE) functional, is widely used for initial structural optimization because it provides an effective balance between computational cost and predictive capability. The total energy of the system is minimized with respect to lattice parameters and atomic coordinates. Structural relaxation continues until predefined convergence criteria are reached for energy and atomic forces.

The optimized lattice should subsequently be examined for physically meaningful bond lengths, octahedral geometry, and structural distortion.

Electronic-Structure Calculations

After structural relaxation, electronic calculations are conducted to determine:

- band structure;
- band-gap energy;
- valence-band characteristics;
- conduction-band characteristics;
- density of states;
- partial density of states;
- orbital contributions near the band edges.

The electronic structure provides essential information about whether the material is semiconducting and whether the electronic states are favorable for photovoltaic charge generation. Because standard PBE calculations can underestimate semiconductor band gaps, a more accurate functional or correction, such as the modified Becke–Johnson (mBJ) approach or a hybrid functional, may be used for improved electronic-structure prediction. Recent computational photovoltaic studies similarly combine conventional DFT with higher-accuracy

approaches when assessing band gaps and optical properties.

Density of States

The total density of states identifies the distribution of electronic states as a function of energy. Partial density of states further determines which atomic orbitals contribute to the valence and conduction bands. For $\text{Rb}_2\text{NaAlI}_6$, particular attention should be given to the contribution of iodine p states and the electronic states associated with Na and Al. Understanding the orbital composition of the band edges can provide insight into optical transitions and carrier transport.

8.5 Optical Properties

The optical properties can be obtained from the calculated frequency-dependent dielectric function.

Important parameters include:

- dielectric constant;
- absorption coefficient;
- refractive index;
- extinction coefficient;
- reflectivity;
- optical conductivity.

The absorption coefficient is especially important because a photovoltaic absorber should efficiently capture incident solar radiation. An enhanced absorption coefficient in the visible region would suggest that relatively thin absorber layers may potentially be sufficient for effective light harvesting.

Structural Stability and Material Suitability

Structural stability represents the first requirement for photovoltaic application. A material may possess a theoretically attractive band gap but remain unsuitable if its crystal structure is energetically or dynamically unstable. The structural assessment of $\text{Rb}_2\text{NaAlI}_6$ should therefore include several complementary criteria. Formation energy can be used to examine thermodynamic favorability relative to competing phases. Tolerance-factor analysis can provide an

initial geometric assessment of the compatibility between ionic sizes. However, tolerance factors should not be treated as definitive evidence of stability because they provide simplified geometric criteria.

More rigorous assessment can include phonon-dispersion calculations and ab initio molecular dynamics. The absence of imaginary phonon modes would provide evidence of dynamic stability. Molecular-dynamics simulations at relevant temperatures can further evaluate whether the crystal structure remains intact under thermal fluctuations.

This multi-criterion approach is consistent with contemporary computational studies of lead-free double perovskites, where structural stability is increasingly evaluated through formation energies, phonons, and molecular dynamics rather than through a single geometric descriptor.

Electronic Properties

The electronic properties of $\text{Rb}_2\text{NaAlI}_6$ are central to its potential photovoltaic application. A suitable absorber should have a band gap that enables substantial solar-spectrum absorption while maintaining sufficient voltage generation.

The calculated band structure should therefore be examined to determine whether the fundamental transition is direct or indirect. Direct-gap materials can provide efficient optical transitions because photons can promote carriers without requiring substantial momentum compensation. The density-of-states analysis should then identify the orbital contributions responsible for the valence-band maximum and conduction-band minimum.

If iodine-derived states dominate the upper valence band, strong halide-related p-orbital contributions may enhance optical transitions. Meanwhile, the chemical identity of the conduction-band states can determine carrier effective masses and transport characteristics.

The electronic structure also provides a foundation for predicting band alignment with charge-transport materials. The conduction-band position should be compatible with electron

extraction, while the valence-band position should permit efficient hole extraction.

Optical Absorption and Solar-Spectrum Utilization

Optical absorption is one of the most important indicators of photovoltaic suitability. A solar-cell absorber must interact strongly with incoming photons and generate electron-hole pairs efficiently. Iodide-based compositions are particularly attractive because iodine has relatively high polarizability and can influence the electronic states associated with optical transitions. Consequently, $\text{Rb}_2\text{NaAlI}_6$ warrants investigation for absorption extending across the visible and potentially into the near-infrared region.

A strong absorption coefficient would provide two major advantages. First, it could enable efficient utilization of incident sunlight. Second, it could reduce the thickness required for the absorber layer.

However, optical absorption alone is insufficient to establish photovoltaic performance. Absorbed photons must generate mobile carriers, and those carriers must reach the interfaces without undergoing severe recombination. Therefore, optical properties must be considered together with carrier lifetime, mobility, defect density, and interface quality. Recent reviews of lead-free perovskites emphasize this interconnected relationship between composition, optical response, electronic structure, stability, and device performance.

Photovoltaic Performance Optimization

The theoretical photovoltaic performance of $\text{Rb}_2\text{NaAlI}_6$ can be optimized through several strategies.

Band-Gap Engineering

The first strategy involves controlling the band gap through chemical substitution. Partial replacement of A-site or halide species could tune the electronic structure. For example, controlled halide substitution could modify lattice parameters and orbital interactions. Similarly, partial cation substitution may provide a route

for adjusting band-edge positions. However, compositional modification must preserve structural stability. Excessive substitution can introduce lattice distortion, phase segregation, or defect formation.

Defect Engineering

Defects can strongly influence photovoltaic performance. Vacancies, antisite defects, interstitials, and substitutional defects can create electronic states inside the band gap. Some defect states can act as recombination centers and reduce carrier lifetime. Therefore, defect suppression and passivation are essential for practical devices. Recent research identifies defect passivation as a major pathway for improving lead-free double-perovskite solar cells.

Interface Engineering

Even a high-quality absorber can perform poorly if the interfaces between the absorber and charge-transport layers are unfavorable. An optimized electron-transport layer should facilitate electron extraction while blocking holes. Conversely, a hole-transport layer should selectively extract holes while minimizing electron leakage. Energy-level alignment is therefore crucial. Computational band-edge analysis can help identify suitable transport materials before device fabrication.

Absorber Thickness

Absorber thickness represents another important optimization variable. A thin absorber may reduce recombination and material consumption but could result in incomplete light absorption. A thicker layer may improve absorption but increase the probability of carrier recombination. Computational device modeling can therefore identify an optimum thickness by simultaneously considering generation and recombination processes.

Proposed Device Architecture

A conceptual $\text{Rb}_2\text{NaAlI}_6$ solar cell can be represented as:

Transparent conducting electrode / Electron transport layer / $\text{Rb}_2\text{NaAlI}_6$ absorber / Hole transport layer / Metal electrode

A suitable transparent conducting electrode provides optical transparency and electrical conductivity. The electron-transport layer should possess favorable conduction-band alignment with $\text{Rb}_2\text{NaAlI}_6$, allowing photogenerated electrons to move efficiently toward the front contact. The $\text{Rb}_2\text{NaAlI}_6$ layer serves as the primary absorber and generates electron-hole pairs following photon absorption. The hole-transport layer facilitates selective extraction of holes toward the rear electrode.

The final metal electrode provides electrical collection.

The precise transport materials should be selected through band alignment calculations and device simulations rather than assumed solely on the basis of commonly used materials.

Device-Level Modeling

To translate the calculated material properties into predicted solar-cell performance, a device simulator such as SCAPS-1D can be integrated into the research framework.

The principal parameters include:

- absorber band gap;
- electron affinity;
- dielectric constant;
- absorber thickness;
- defect density;
- carrier mobility;
- electron and hole effective masses;
- interface defect density;
- transport-layer properties;
- temperature;
- illumination intensity.

The principal photovoltaic outputs include:

- open-circuit voltage (V_{oc});
- short-circuit current density (J_{sc});
- fill factor (FF);
- power conversion efficiency (PCE).

The relationship can be expressed as:

$$\text{PCE} = V_{oc} \times J_{sc} \times \text{FF} \times 100 \quad \text{PCE} = \frac{V_{oc} \times J_{sc} \times \text{FF}}{P_{in}} \times 100$$

where P_{in} represents the incident solar power density.

For standard AM1.5G illumination, P_{in} is conventionally taken as approximately 100 mW cm^{-2} .

Device-level optimization should initially use idealized parameters derived from first-principles calculations and subsequently introduce increasingly realistic defect and interface conditions.

Expected Performance Relationships

The computational framework predicts several important relationships.

First, reducing an excessively large band gap would increase the fraction of the solar spectrum available for absorption and could increase J_{sc} .

Second, improved carrier mobility would reduce transport losses and potentially increase the fill factor.

Third, reduced defect density would suppress nonradiative recombination and improve both V_{oc} and FF.

Fourth, appropriate band alignment would improve carrier extraction and reduce interfacial recombination.

Fifth, optimized absorber thickness would balance photon absorption against carrier recombination.

These relationships demonstrate why photovoltaic optimization must move beyond the simple identification of a theoretically favorable band gap.

Discussion

The significance of $\text{Rb}_2\text{NaAlI}_6$ lies not simply in its lead-free composition but in the possibility of engineering its electronic and optical characteristics through chemical and structural design. The current generation of lead-free perovskite research demonstrates that eliminating lead is necessary but does not automatically produce a high-performance photovoltaic

material. Some lead-free compositions possess excellent stability but unfavorable band gaps, while others demonstrate strong optical absorption but suffer from poor charge transport or substantial defect-mediated recombination. This trade-off has been emphasized by recent reviews. Ahmed et al. (2024) reported that although numerous lead-free systems have emerged, substantial technical challenges remain before these materials can compete effectively with mature lead-based perovskite technologies. Similarly, Miah et al. (2025) highlighted the importance of simultaneously addressing toxicity, stability, and photovoltaic performance when developing sustainable perovskite technologies. The computational strategy adopted in the present study addresses this challenge by connecting structural characteristics with electronic and optical properties.

Structural–Electronic Relationship

Structural parameters can significantly influence orbital overlap and electronic dispersion. Changes in lattice constants modify bond lengths and bond angles, which subsequently affect electronic states. For double perovskites, chemical ordering between B and B' cations is particularly important because the electronic band edges can originate from different atomic orbitals. This provides opportunities for deliberate electronic-structure engineering.

Optical–Electronic Relationship

The optical response is closely related to the electronic band structure. A smaller band gap generally permits absorption of lower-energy photons, while stronger allowed transitions can enhance absorption coefficients. Therefore, the suitability of $\text{Rb}_2\text{NaAlI}_6$ should be evaluated not only through its fundamental band gap but also through the calculated dielectric function and absorption coefficient.

Stability–Performance Relationship

A photovoltaic material must remain stable while exposed to heat, light, electric fields, and

environmental conditions. Lead-free materials are not automatically stable under all operating conditions. Moisture, oxygen, thermal stress, illumination, and defects can cause structural degradation. Recent research therefore increasingly emphasizes a multi-dimensional definition of stability incorporating thermodynamic, dynamic, thermal, chemical, and operational stability.

Environmental and Sustainability Considerations

The environmental motivation for investigating $\text{Rb}_2\text{NaAlI}_6$ is closely associated with the broader movement toward lead-free photovoltaics. Lead contamination remains a major concern because photovoltaic modules are intended for long operational lifetimes and large-scale deployment. Even if encapsulation reduces exposure during operation, end-of-life management remains important. Lead-free double perovskites potentially reduce this environmental burden. However, sustainability should not be evaluated solely by excluding lead. The toxicity, abundance, extraction requirements, recyclability, and environmental persistence of all constituent elements should be considered. Therefore, future research on $\text{Rb}_2\text{NaAlI}_6$ should include life-cycle assessment, material availability analysis, recycling pathways, and environmental-impact evaluation.

Limitations of the Study

Several limitations should be recognized.

First, DFT calculations generally describe ideal crystalline structures. Real materials contain grain boundaries, vacancies, impurities, antisite defects, and structural disorder.

Second, conventional exchange-correlation functionals may underestimate semiconductor band gaps. Consequently, hybrid functionals or mBJ calculations should be considered for improved accuracy.

Third, theoretical optical properties do not automatically reproduce experimental thin-film behavior. Grain boundaries and processing-

related defects can significantly modify optical and electrical properties.

Fourth, predicted photovoltaic efficiency depends strongly on assumed defect densities, carrier mobilities, interface properties, and layer parameters.

Finally, computational predictions require experimental validation. Synthesis, structural characterization, optical measurements, and device fabrication are necessary to establish the practical potential of $\text{Rb}_2\text{NaAlI}_6$.

Future Research Directions

Future research should focus on five major areas.

Experimental Synthesis

The first priority should be the experimental synthesis of phase-pure $\text{Rb}_2\text{NaAlI}_6$. X-ray diffraction can be used to confirm crystal structure, while scanning electron microscopy can assess morphology.

Advanced Electronic Calculations

Hybrid-functional and many-body calculations should be used to obtain more accurate band gaps and optical transitions.

Defect Physics

Formation energies of vacancies and antisite defects should be calculated under different chemical potentials. This would provide important information regarding intrinsic defect tolerance.

Device Simulation

SCAPS-1D or comparable simulation platforms can be used to systematically optimize absorber thickness, defect density, transport-layer thickness, interface recombination, and temperature.

Machine-Learning-Assisted Optimization

Machine learning could eventually be combined with DFT calculations to identify promising compositional modifications of $\text{Rb}_2\text{NaAlI}_6$. Recent computational reviews identify the integration of DFT and data-driven approaches as

an increasingly important direction in lead-free perovskite research.

Conclusion

This study presents a computational framework for investigating and optimizing $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double-perovskite solar cells. The research is motivated by the need to develop photovoltaic materials that combine favorable optoelectronic properties with improved environmental sustainability.

The proposed methodology integrates crystal-structure optimization, density functional theory, electronic-structure analysis, density-of-states calculations, optical-property evaluation, and device-level optimization. Such an integrated strategy is essential because photovoltaic performance depends on the interaction of structural, electronic, optical, and interfacial properties.

$\text{Rb}_2\text{NaAlI}_6$ represents a promising composition for further investigation because its iodide-based double-perovskite framework provides a potentially useful platform for optical and electronic engineering. Nevertheless, its practical photovoltaic potential must be established through systematic evaluation of structural stability, band-edge characteristics, optical absorption, defect behavior, and interface compatibility.

The study further demonstrates that material discovery should not rely exclusively on band-gap calculations. Defect suppression, interface engineering, absorber thickness, charge extraction, and carrier recombination must also be incorporated into the optimization process.

Overall, computational investigation of $\text{Rb}_2\text{NaAlI}_6$ can provide an efficient first step toward the development of sustainable photovoltaic materials. Experimental synthesis and characterization should constitute the next stage of research, followed by device fabrication and performance validation. More broadly, the integration of first-principles calculations, device simulation, defect engineering, and machine-learning-assisted materials discovery could

accelerate the development of efficient and environmentally responsible lead-free double-perovskite solar cells.

REFERENCES

Ahmed, S., Gondal, M. A., Alzahrani, A. S., et al. (2024). Recent trends and challenges in lead-free perovskite solar cells: A critical review. *ACS Applied Energy Materials*, 7(4), 1382–1397.

<https://doi.org/10.1021/acsaem.3c02327>

Fatima, S. K., Alzard, R. H., Amna, R., Alzard, M. H., Zheng, K., & Abdellah, M. (2025). Lead-free perovskites for next-generation applications: A comprehensive computational and data-driven review. *Materials Advances*, 6, 7634–7661. <https://doi.org/10.1039/D5MA00681C>

Gao, S., & Bae, J.-J. (2025). Toward sustainable perovskite solar cells: From lead-free materials to environmental concerns and mitigation strategies. *EcoMat*, 7(3), e70001. <https://doi.org/10.1002/eom2.70001>

Khan, A. D., Mustajab, M., Moeen, S., et al. (2024). Advancements in the stability, protection and lead-free strategies of perovskite solar cells: A critical review. *Environmental Science: Advances*, 3, 1004–1029. <https://doi.org/10.1039/D3VA00401E>

Kumar, V., Kathiravan, A., & Jhonsi, M. A. (2024). Beyond lead halide perovskites: Crystal structure, bandgaps, photovoltaic properties and future stance of lead-free halide double perovskites. *Nano Energy*, 125, 109523.

Miah, M. H., Khandaker, M. U., Hossen, M. J., Noor-E-Ashrafi, Jahan, I., Shahinuzzaman, M., Nur-E-Alam, M., Hanfi, M. Y., Ullah, M. H., & Islam, M. A. (2025). Lead-free alternatives and toxicity mitigation strategies for sustainable perovskite solar cells: A critical review. *Materials Advances*, 6, 2718–2752. <https://doi.org/10.1039/D5MA00010F>

Phonglamjiakngam, P., Hazaea, Z. A. M., Wang, M., & Pootrakulchote, N. (2026). Recent progress on raising the efficiency of lead-free double perovskite solar cells. *Solar Energy*, 303, 114164. <https://doi.org/10.1016/j.solener.2025.114164>

Javed, H., Khan, A. N., Khan, N. U., Noubigh, A., Iqbal, M. M. A., & Gul, N. (2026). Materials-to-device assessment of lead-free Na₂BAlI₆ (B = K, Rb) double perovskites for high-efficiency photovoltaics. *Chemical Physics*, 610, 113344. <https://doi.org/10.1016/j.chemphys.2026.113344>