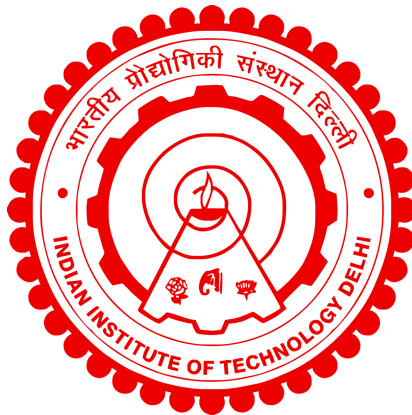


FIRST PRINCIPLES EXPLORATION OF ELECTRONIC PROPERTY MODULATION IN LOW-DIMENSIONAL MATERIALS FOR ENERGY APPLICATIONS

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ENGINEERING

INDIAN INSTITUTE OF TECHNOLOGY DELHI

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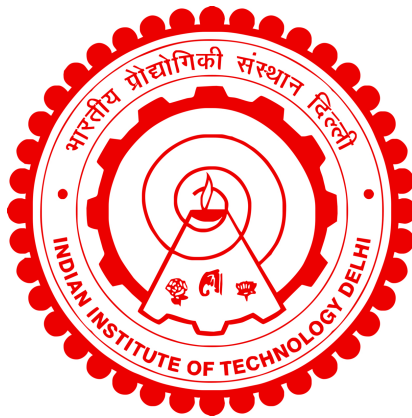
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Department of Materials Science and Engineering

Submitted

in fulfillment of the requirements of the degree of Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

JUNE 2024

Dedicated to My Grandparents

Certificate

This is to certify that the thesis entitled “**First Principles Exploration of Electronic Property Modulation in Low-Dimensional Materials for Energy Applications**”, submitted by **Kiran Yadav** to the Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy** in Materials Science and Engineering department, is a record of the original, bonafide research work carried out by her under my supervision and guidance. The thesis has reached the standards fulfilling the requirements of the regulations related to the award of the degree.

The results contained in this thesis have not been submitted in part or in full to any other University or Institute for the award of any degree or diploma to the best of our knowledge.

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Kiran Yadav

Abstract

Low-dimensional materials, where the motion of charge carriers is confined in one or more dimensions, typically at the scale of the de-Broglie wavelength, possess unique quantum effects and increased surface-to-volume ratios, and have emerged as competitive options for energy-related devices. The role of dimensionality has been explored in a vast range of materials ranging from perovskite materials driving solar cell applications to lightweight conducting materials for portable energy storage. This thesis is divided into two main parts, with an overarching goal of contributing valuable insights into the interplay of dimensionality and electronic properties using density functional theory (DFT).

The first half explores the structure and electronic properties of aluminene, a two-dimensional layer of Aluminium. While two-dimensional (2D) semiconductors have received a lot of attention, the prospect of creating 2D metallic structures has picked up pace recently owing to theoretical and experimental studies in the area. We investigate the structure and stability of 2D aluminene sheets and nanoribbons, crystallizing in a honeycomb like lattice, and explore its potential application as anodes for metal-ion batteries and also for hydrogen storage. The second half of the thesis shifts its focus to low-dimensional perovskite materials, characterized by the essential ABX_3 structural archetype. These materials play a crucial role in various technologically significant properties such as ferroelectricity, piezoelectricity, superconductivity, and photochemical behavior. The soft nature of the perovskites can easily be modulated by the application of strain, and we explore the coupling between dimensionality and strain. Our findings would contribute to the advancement of sustainable technologies and the development of more efficient and resilient energy materials and solutions.

सार

निम्न-आयामी सामग्री, जहाँ चार्ज वाहकों की गति एक या अधिक आयामों में सीमित होती है, आमतौर पर डी-ब्रोगली तरंग दैर्ध्य के पैमाने पर, अद्वितीय क्वांटम प्रभाव और बढ़े हुए सतह-से-आयतन अनुपात रखते हैं, और ऊर्जा-संबंधी उपकरणों के लिए प्रतिस्पर्धी विकल्प के रूप में उभरे हैं। आयामीयता की भूमिका को व्यापक रूप से विभिन्न सामग्रियों में खोजा गया है, जो सौर सेल अनुप्रयोगों को संचालित करने वाली पेरोव्स्काइट सामग्रियों से लेकर पोर्टेबल ऊर्जा भंडारण के लिए हल्के चालक सामग्री तक हैं। यह शोध प्रबंध दो मुख्य भागों में विभाजित है, जिसका मुख्य उद्देश्य घनत्व कार्यात्मक सिद्धांत (DFT) का उपयोग करके आयामीयता और इलेक्ट्रॉनिक गुणों के बीच के संबंध में महत्वपूर्ण अंतर्दृष्टि प्रदान करना है।

पहला भाग एल्युमिनीन, एल्यूमिनियम की द्वि-आयामी परत की संरचना और इलेक्ट्रॉनिक गुणों की जांच करता है। जबकि द्वि-आयामी (2D) अर्धचालकों ने काफी ध्यान आकर्षित किया है, द्वि-आयामी धातु संरचनाएँ बनाने की संभावना हाल ही में इस क्षेत्र में सैद्धांतिक और प्रायोगिक अध्ययनों के कारण गति पकड़ रही है। हम 2D एल्युमिनीन शीट्स और नैनोरिबन की संरचना और स्थिरता की जांच करते हैं, जो एक मधुकोश जैसी जाली में क्रिस्टलीकृत होते हैं, और इसे धातु-आयन बैटरियों के लिए एनोड और हाइड्रोजन भंडारण के लिए संभावित अनुप्रयोग के रूप में इसके संभावनाओं का पता लगाते हैं।

शोध प्रबंध का दूसरा भाग निम्न-आयामी पेरोव्स्काइट सामग्रियों पर ध्यान केंद्रित करता है, जिनकी विशेषता आवश्यक ABX_3 संरचनात्मक आर्किटाइप है। इन सामग्रियों का विभिन्न तकनीकी रूप से महत्वपूर्ण गुणों जैसे फेरैइलेक्ट्रिसिटी, पाईजोइलेक्ट्रिसिटी, सुपरकंडक्टिविटी, और प्रकाश रासायनिक व्यवहार में महत्वपूर्ण भूमिका होती है। पेरोव्स्काइट्स की कोमल प्रकृति को आसानी से तनाव के अनुप्रयोग द्वारा मॉड्यूलेट किया जा सकता है, और हम आयामीयता और तनाव के बीच के युग्मन की खोज करते हैं। हमारे निष्कर्ष स्थायी प्रौद्योगिकियों की प्रगति और अधिक कुशल और लचीली ऊर्जा सामग्री और समाधानों के विकास में योगदान देंगे।

Contents

Certificate

Acknowledgements

Abstract

Contents

List of Figures

List of Tables

1	Introduction	1
1.1	Overview of low-dimensional materials for energy conversion and storage	4
1.1.1	Two-dimensional materials	4
1.1.1.1	Synthesis Techniques for Metallenes	7
1.1.2	Low dimensional halide perovskites	10
1.1.2.1	Synthesis of low-dimensional perovskites	12
1.1.2.2	Strain Effects on Metal Halide Perovskites	15
1.1.3	Other Nanostructured materials	16
1.2	Advantages and challenges of low-dimensional materials for energy conversion and storage	18
1.2.1	Solar cell applications	19
1.2.2	Metal-ion batteries	20
1.2.3	Supercapacitors	21
1.2.4	Hydrogen storage	22
1.3	Thesis overview	23
2	Computational Methodology	25
2.1	First principles calculations	26
2.1.1	Overview of approximations	27
2.1.2	Density-functional theory	28
2.1.2.1	Kohn-Sham equations	30

2.1.3	Exchange correlation functionals	31
2.1.4	Electron-ion interactions and pseudopotential approximation	32
2.1.5	Periodic supercells	33
2.1.5.1	Low dimensional and aperiodic systems	34
2.2	Quantum Transport for Nanoscale Systems	34
2.3	Computational implementation and softwares	36
3	Structure and electronic properties of low dimensional metallic Aluminium	39
3.1	Background and motivation for the study	39
3.2	Computational details	41
3.3	Free standing aluminene	42
3.4	Aluminene nanoribbons	46
3.4.1	Passivation with O and H atoms	48
3.5	Transport through low dimensional aluminene	49
3.5.1	Pristine and single point defect aluminene	49
3.5.2	Passivated and unpassivated nanoribbons	52
3.6	Conclusion	54
4	Dual-Duty Innovation: Two-dimensional Aluminum Anodes for Metal Ion Batteries and Promising Prospects for Hydrogen Storage	57
4.1	Background and motivation for the study	58
4.2	Computational Methods	60
4.3	2D aluminene for metal-ion batteries	61
4.3.1	Adsorption of metal atoms on aluminene	61
4.3.2	Migration of metal atoms on aluminene	66
4.3.3	Open circuit voltage and maximum theoretical storage capacity	68
4.4	Potential of 2D aluminene for hydrogen storage	73
4.4.1	Hydrogen adsorption on pristine Al monolayer	73
4.4.2	Hydrogen adsorption on Li-decorated Al monolayer	77
4.4.3	Thermodynamic analysis	83
4.4.4	Comparison and prospect of hydrogen storage capacities	85
4.5	Conclusion	88
5	2D metal halide perovskites for solar cell applications: Modulation of electronic properties through surface termination and biaxial strain	91
5.1	Background and motivation for the study	92
5.2	Computational details	93
5.3	2D Cs ₂ BCl ₄ & CsB ₂ Cl ₅ (B = Pb,Sn): Structure and stability	96
5.3.1	Effect of spin-orbit coupling	100

5.3.2	Thermodynamic analysis	101
5.3.3	Effect of biaxial strain	104
5.4	Effect of layer thickness	108
5.4.1	Comparison with the effect of triaxial strain in 3D perovskites	112
5.5	Implications for solar cell applications	115
5.6	Conclusion	120
6	Picoperovskites : Electronic properties of 1D metal halide perovskites	123
6.1	Background and motivation for the study	123
6.2	Computational details	125
6.3	Structure and electronic properties of 1D perovskite and perovskite-like structures	127
6.3.1	Effect of compressive and tensile strain	130
6.3.2	Effect of carbon nanotube encapsulation	131
6.4	Conclusion	134
7	Summary and Future Work	137
	References	141
	List of Publications	183
	Curriculum Vitae	185

List of Figures

1.1	Illustrating the versatile energy-related fields which benefit significantly from the roles played by nanomaterials, with thermoelectric, piezoelectric, triboelectric, photovoltaic, catalytic, and electrochromic nanomaterials emerging as pivotal contributors.	3
1.2	(a) Summary of metallene positions in the periodic table, obtained through either experimental synthesis or theoretical predictions. Metallene classification is determined by monolayer for main group metals and few layers (< 5 nm) for transition metals. (b) Wheel-shaped schematic representation featuring key applications in the energy sector, accompanied by a timeline showcasing the experimental milestones of diverse metallenes. The designation <i>Th.</i> denotes theoretical studies.	6
1.3	(a) Comparison of the lateral size versus aspect ratio (thickness-to-lateral scale) for the 2D metals synthesized by different routes discussed in the main text. Here, h represents the thickness. (b) Images of various 2D metals realized experimentally. Low-voltage spherical aberration-corrected transmission electron microscopy (TEM) image of a monoatomic Fe layer with the square unit cell suspected in a graphene pore. Fabrication process of Bi nanosheets (BiNSs) using pristine Bi nanoparticles via hot-pressing method, and atomic force micrographs of resulting BiNSs on Si/SiO ₂ substrate. Atomic resolution scanning tunnelling micrograph of ~ 5 layers 2D-Sb. SEM and HRTEM images of 2D Ti obtained by polymer surface buckling enabled exfoliation (PSBEE) suspended over the copper grid for observation.	8
1.4	Schematic showing possible morphological low-dimensional forms of ABX ₃ halide perovskites with key advantages and possible applications.	11

1.5	Various methods employed in the creation of low-dimensional halide perovskites and images of synthesized structures. (a) 2D halide perovskites; (b) 1D halide perovskites; (Top row, left to right) Schematic representations of three CsPbX_3 orthorhombic crystal structures; TEM images of CsPbBr_3 and CsPbI_3 nanowires. (Bottom row, left to right) TEM and aberration-corrected HRTEM images of CsPbBr_3 ultrathin nanowires; TEM image of the CsSnI_3 nanorods. (c) 0D halide perovskites; Schematic of the cubic CsPbBr_3 structure and TEM image of CsPbBr_3 nanocrystals synthesized using hot-injection method; Normalized photoluminescence (PL) spectra of CsPbX_3 nanocrystals with different compositions.	14
1.6	Illustration of the diverse range of nanoscale building blocks for creating advanced energy storage devices, this figure showcases their chemical, structural, and morphological diversity. It highlights potential improvements over current devices while summarizing the advantages and challenges associated with each class of nanomaterials in the final rows. Image taken from [1].	17
1.7	Absorption spectra of PbSe quantum dots spanning the range from 3.3 nm to 8.1 nm . Several discrete transitions are observable and represent discrete excitonic transitions. Top right depicts the increased quantum confinement that the 1st exciton experiences as the particle size decreases. Bottom right shows variation of bandgap E_g , as estimated from the lowest energy transition as a function of the dot radius.	19
2.1	Multi-scale materials simulation at various length and time scales. . .	26
2.2	Schematic representation of pseudopotential approximations. Ionic potential and valence wave function is shown by green color, and the corresponding pseudopotential and pseudo wavefunction is shown using blue color.	33
2.3	Schematic showing (a) two-dimensional (2D) graphene sheet, (b) one-dimensional (1D) carbon nanotube and (c) zero-dimensional (0D) fullerene structures.	35
2.4	A schematic illustration of two-terminal device geometry showing semi-infinite electrodes and finite scattering region including channel and connecting leads.	37
2.5	Flowchart of DFT loop	38

3.1	Visualization of top and side view of hexagonal honeycomb aluminene in (a) planar and (e) buckled structure. Phonon dispersion spectra of (b) planar and (f) buckled structure, highlighting relative stability of buckled structure. Bandstructure and projected density of states (PDOS) of (c,d) planar and (g,h) buckled structure indicating metallic behaviour of aluminene. The Fermi level (indicated by dotted solid line) has been set to 0.	43
3.2	Visualisation of aluminene nanoribbons and their corresponding band structure in various configuration (a,b) 3p Armchair, (c,d) 3p+1 Armchair, (e,f) 3p+2 Armchair and (g,h) Zigzag configuration. Fermi level has been set to zero and depicted using dashed line for all.	45
3.3	Visualisation of H-atom passivated aluminene nanoribbons and their corresponding band structure in various configuration (a,b) 3p Armchair, (c,d) 3p+1 Armchair, (e,f) 3p+2 Armchair and (g,h) Zigzag configuration. The Fermi level has been set to zero.	47
3.4	Visualisation of O-atom passivated aluminene nanoribbons and their corresponding band structure in various configuration (a,b) 3p Armchair, (c,d) 3p+1 Armchair, (e,f) 3p+2 Armchair and (g,h) Zigzag configuration. Fermi level has been set to zero and depicted using dashed line for all. The Fermi level crossing by bands highlights metallicity of passivated nanoribbons.	50
3.5	(a) Schematic of the armchair and zig-zag directions in the 2D sheet and corresponding electrode and scattering regions for transport calculations. (b) Transmission (in channels/Å for armchair and zigzag directions of the 2D sheet. (c) Current (normalized by unit cell width) as a function of applied voltage, showing Ohmic behaviour at low voltages.	51
3.6	(a) Schematic of the defect-free and (b) vacancy point defect 2D aluminene sheet and corresponding electrode and scattering regions for transport calculations. (c) Transmission (in channels/Å for defect-free and defected 2D sheet along zigzag direction. (d) Current (normalized by unit cell width) as a function of applied voltage, showing decrease in current in vacancy defect sheet due to reduced number of channels.	53
3.7	(a) Schematic of the unpassivated and H-atom passivated aluminene nanoribbons and their corresponding electrode and scattering regions for transport calculations. (b) Conductance quantization in nanoribbons.	54

4.1	(a) Unit cell of buckled aluminene monolayer with symmetry elements of underlying $p3\bar{m}1$ layer group as described in International Tables for Crystallography Volume E: Subperiodic groups. The two Al atoms shaded differently to indicate the buckling of the atoms relative to each other, occupy Wyckoff position 2c with site symmetry 3m. (b) Supercell showing the high symmetry adsorption sites: B (square), D (circle), H (triangle), T (inverted triangle), and X (rhombus). (c) Adsorption energy, E_{ads} in eV for adsorption of metal atoms at each of the indicated high symmetry sites.	62
4.2	Phonon dispersion spectra of the 2×2 (a) aluminene supercell (b) aluminene supercell with Li atom adsorbed (c) aluminene supercell with Na atom adsorbed, indicating the dynamical stability of the aluminene before and after adsorption.	63
4.3	(a) Projected density of states (PDOS) showing Al and M atom states contribution around the Fermi level states, (b) electron localization function (ELF) visualizations for adsorption of Li, Na, K, Mg and Ca, and (c) Charge density difference visualizations for different adatoms as indicated. The yellow (blue) regions indicate charge density accumulation (depletion) regions. Arrows indicate the Bader charge transfer from the adatom to the monolayer.	65
4.4	(a) Schematic illustrating the diffusion path of D-site to D-site for Li and Na, and (b) T-site to T-site for K and Ca. (c) Diffusion barriers, E_b for the indicated pathways for Li (pink squares), Na (blue circles), K (green triangles) and Ca (blue circles). (d) Barrier heights, E_b , in eV for Li (pink), Na (blue), K (green) and Ca (red).	67
4.5	(a) Schematic of Na penetration through the aluminum monolayer thickness, taking place via H-site or the honeycomb ring center. (b) Profiles of energy barrier, E_b (in eV) encountered by the K (green), Na (blue), Ca (red) and Li (pink) metal atom during diffusion.	68
4.6	Top and side view of 2D aluminum depicting structural changes with coverage of up to 1 monolayer metal atoms on it. Pink and blue represent Li and Na atoms, respectively.	70
4.7	(a) Adsorption energy, E_{ads} as a function of increasing Li (pink squares) and Na (blue circles) concentration for single side and (b) double side coverage. N_M denotes the number of metal atoms adsorbed (c) Open circuit voltage (OCV) as a function of increasing Li (pink squares) and Na (blue circles) concentration, within the double side coverage model. (d) Comparison of the average OCV as a function of maximum reported capacity, C , for this work with literature reports on other elemental 2D materials - β_{12} -borophene, T-graphene, phosphorene, silicene, germanene, stanene, arsenene, bismuthene and antimonene. Squares and circles symbols represent values for Li and Na respectively.	72

4.8	(a) Charge density difference visualizations with H_2 molecule adsorbed at D-site of aluminene, with arrows indicating charge transfer direction. The yellow area represents the electron accumulation, indicating an increase of charge density, and the cyan area represents the electron loss, indicating a decrease of charge density. (b) Projected density of states (PDOS) of pristine aluminene showing contribution of Al s- and p-orbitals near the Fermi level.	73
4.9	Minimum energy pathways and energetics of diffusion through the H-site of the monolayer for (a) single hydrogen atom, (b) single hydrogen molecule approaching perpendicular to the surface and (c) single hydrogen molecule approaching parallel to the surface. Insets show visualizations for the initial, transition state in the middle and the final states. Red and Grey color atoms represents H-atom and Al-atom, respectively.	76
4.10	(a) Schematic of aluminene with Li-decoration sites marked as D and T. (b) Top and side view visualizations of charge density difference of decorated aluminene. The yellow area represents the electron accumulation, indicating an increase of charge density, and the cyan area represents the electron loss, indicating a decrease of charge density. Arrow indicating charge transfer from Li to aluminene. (c) Projected density of states after Li-adsorption on aluminene showing contribution of Li atom s and p states around fermi level, E_f . (d) Adsorption energy as a function of varied H_2 molecule numbers with Li-decoration at D, H and T-site.	78
4.11	Topview and sideview of Charge density difference visualizations of single Li-decorated (at T-site) aluminene with (a) single H_2 , (b) $2H_2$, (c) $3H_2$ and $6H_2$ molecules. The yellow area represents the electron accumulation area, indicating an increase of charge density, and the cyan area represents the electron loss area, indicating a decrease of charge density. (e-h) Plots shows H and Li s-orbital projected density of states (PDOS) for $1H_2$, $2H_2$, $3H_2$, and $6H_2$ molecules adsorption on Li-decorated aluminium monolayer. We use H_t and H_d for the top and bottom H-atoms of vertical H_2 molecule.	82
4.12	E_r as a function of (a) temperature with pressure taken as standard 0.1MPa (b) pressure with temperature taken as $RT = 298.15$ K, for single H_2 , 2 H_2 and 3 H_2 molecules adsorbed at T-site Li decorated aluminene.	85
5.1	Side view of (a) 2D Cs_2BCl_4 and (b) CsB_2Cl_5 structures. Pink arrows denote rumpling in the structure. Top view of charge density isosurfaces expanded on 4×4 supercells of (c) 2D Cs_2BCl_4 and (d) CsB_2Cl_5	97

5.2	Electronic band structure along the high symmetry path $\Gamma \rightarrow M \rightarrow X$ and total and orbital resolved density of states for strain-free (a) Cs_2PbCl_4 , (b) Cs_2SnCl_4 , (c) CsPb_2Cl_5 and (d) CsSn_2Cl_5 . The Fermi level, E_f , has been set to zero in all cases.	99
5.3	Band dispersion and density of states (DOS) of bulk cubic CsBCl_3 (B=Pb,Sn) perovskites with and without spin-orbit coupling (SOC) influence. We use solid lines for with SOC and dotted lines without spin-orbit coupling (SOC). For all systems, valence band maxima (VBM) has been set to zero. The blue and green curves correspond to Pb and Sn, respectively.	101
5.4	Phase diagrams for (001) surfaces of (a) CsPbCl_3 and (b) CsSnCl_3 . Shaded regions highlight the termination that is most stable in relation to the chemical potentials of Cs and Cl, where yellow corresponds to CsCl termination, and blue and green represent PbCl_2 and SnCl_2 terminations respectively. The conditions for thermodynamically stable CsSnX_3 bulk phase are highlighted by the red dashed lines. (c,d) Surface energies (E_{surf}) of Cs_2BCl_4 (solid lines) and CsB_2Cl_5 (dotted lines) (B=Pb, Sn) surfaces as a function of $\Delta\mu_{Cl}$. The conditions of stable CsPbCl_3 and CsSnCl_3 are marked by blue and green areas. . .	103
5.5	(a) Schematic showing the direction of tensile (hollow arrows) and compression (filled arrows) strains on the 2D perovskites. (b) Formation energy, E_{form} computed from binary phase energies. Blue squares represent Cs_2PbCl_4 , green squares represent Cs_2SnCl_4 , blue triangles represent CsPb_2Cl_5 , and green triangles represent CsSn_2Cl_5 . (c) Rumpling (in Å) and electronic bandgap, and (d) E_g as a function of applied biaxial strain, ϵ_b . Blue squares represent Cs_2PbCl_4 , green squares represent Cs_2SnCl_4 , blue triangles represent CsPb_2Cl_5 , and green triangles represent CsSn_2Cl_5	106
5.6	Effective mass, m^* variations with strain, along $M \rightarrow \Gamma$ and $M \rightarrow X$ for (a) 2D Cs_2PbCl_4 and (b) 2D Cs_2SnCl_4 . Hollow and filled markers have been used for holes and electrons, respectively. Direction average mobility variations with biaxial strain for (c) Cs_2PbCl_4 and (d) Cs_2SnCl_4	107
5.7	Effective mass, m^* variations with strain, along $M \rightarrow \Gamma$ and $M \rightarrow X$ for bilayer (a) 2D $\text{Cs}_3\text{Pb}_2\text{Cl}_7$ and (b) 2D $\text{Cs}_3\text{Sn}_2\text{Cl}_7$. Hollow and filled markers have been used for holes and electrons, respectively. Direction average mobility variations with biaxial strain for (c) $\text{Cs}_3\text{Pb}_2\text{Cl}_7$ and (d) $\text{Cs}_3\text{Sn}_2\text{Cl}_7$	110
5.8	Effective mass, m^* variations with strain, along $R \rightarrow \Gamma$, $R \rightarrow X$ and $R \rightarrow M$ for (a) CsPbCl_3 and (b) CsSnCl_3 Bulk. Hollow and filled markers have been used for holes and electrons, respectively. Direction average mobility variations with biaxial strain for (c) CsPbCl_3 and (d) CsSnCl_3	111

5.9	(a) Schematic showing bulk, bilayer and monolayer structures with (001) CsCl termination. Each structure is characterized by $1/N$, where N denotes the number of unit cells along the z -axis. (b) Electronic bandgap, E_g as a function of $1/N$ for Pb and Sn systems. Variation of (c) direction average electron and hole mobility, and (d) percentage mobility variation with 2% compressive strain with thickness ($1/N$). Hollow and filled markers have been used for holes and electrons, respectively.	113
5.10	Variation of effective mass, m^* , and corresponding mobility, μ , with applied triaxial strain, along $R \rightarrow \Gamma$, $R \rightarrow X$, and $R \rightarrow M$ for (a,d) CsPbCl ₃ , (b,e) CsSnCl ₃ . Hollow and filled markers have been used for holes and electrons, respectively.	114
5.11	(a) Static dielectric constant, ϵ and (b) exciton binding energy as a function of applied triaxial strain. Blue, green, and red curves correspond to Pb, Sn, and Ge, respectively.	116
5.12	Polarization given Born effective charges, Z^* variation with strain and B-atom change of CsBCl ₃ for (a) Cs-atom, (b) B-atom, and (c,d) Cl-atoms. Blue, green, and red curves correspond to CsPbCl ₃ , CsSnCl ₃ , and CsGeCl ₃ , respectively.	118
6.1	A. Schematic model of 1D perovskite nanowires confined in single-walled carbon nanotubes (SWCNT). Red: lead atoms; orange: iodine atoms; blue: cesium atoms B-D. ADF-STEM characterization of several-single unit cell thick Cs-Pb-I perovskite nanowires. E. HRTEM image of CsSnI ₃ bilayer structure F. Unoptimized and DFT-optimized structures for bilayer structure in a (9,9) SWCNT.	124
6.2	Schematic representation of single unit-cell thick (a) CsPbX ₃ and (b) Cs ₄ PbX ₅ . Bandstructures for 1D CsPbX ₃ for (c) X=Cl, (e) X=Br, (g) X=I, and 1D Cs ₄ PbX ₅ for (d) X=Cl, (f) X=Br, (h) X=I. Green, Brown and Violet color have been used for Cl, Br and I halide respectively.	128
6.3	(a) Schematic showing application of uniaxial strain. Solid arrow marker depicts compression and hollow arrow depicts dilation. (b) Bandgap variation with strain. (c) Hole and (d) electron mobility variation with strain. Green, Brown and Violet color have been used for Cl, Br and I halide respectively.	129
6.4	Top and sideview of charge density difference visualization for Cs ₄ PbI ₅ encapsulated in (a) armchair and (b) zigzag SWCNT, and CsPbI ₃ encapsulated in (a) armchair and (b) zigzag SWCNT.	131
6.5	Total and resolved DOS for Cs ₄ PbI ₅ encapsulated in (a) armchair and (b) zigzag SWCNT, and CsPbI ₃ encapsulated in (c) armchair and (d) zigzag SWCNT.	132

6.6	(a) Distribution of C-C bond lengths along the diameter of the encapsulating zigzag CNT, when bond lengths are fixed (blue), and when allowing for distortions (green) in the CNT structure due to the Cs_4PbI_5 nanowire. (b) Density of states for the fixed and relaxed Cs_4PbI_5 encapsulated in zigzag nanotube.	133
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List of Tables

3.1	Formation energies for pristine and edge passivated aluminene nanoribbons.	48
4.1	Adsorption energies of H_2 molecule at different sites when placed parallel and perpendicular to the aluminium monolayer.	75
4.2	Comparing H_2 molecule adsorption energy (E_{ads}), H-H bond length (H-H), H_2 molecule adsorption heights from monolayer surface (h_{H_2Al}), distance between alkali-atom(M) and H_2 molecule (d_{H_2M}), and adsorption height alkali metal from aluminium surface (h_{MAI}) etc. with change of alkali atom decoration. We use most stable adsorption site of alkali metal on aluminium for H_2 adsorption.	79
4.3	Comparison of average adsorption energy per H_2 molecule (E_{ads}), H_2 molecule bond length (H-H), H_2 molecule adsorption heights from monolayer surface (h_{H_2-Al}), distance between Li and H_2 molecule (d_{H_2-Li}), and adsorption height of Li metal from aluminium surface (h_{Li-Al}) etc. with change of Li-decoration site.	80
4.4	Comparison of Li decorated 2D monolayers gravimetric hydrogen storage capacities (wt %), average adsorption energies per H_2 molecule (E_{ads}), and the maximum number of hydrogen molecules a single Li-atom can adsorb ($N_{max}H_2/Li$).	86
5.1	Comparison of computed and experimentally observed lattice parameters and bandgaps for bulk cubic $CsPbCl_3$ without the inclusion of spin-orbit coupling.	96
5.2	Comparison of structural parameters of 2D monolayer perovskites. . .	98
5.3	Comparison of structural and electronic parameter evolution with thickness and strain.	112
5.4	Born effective charges, (Z^*) values	119
6.1	Bandgap, E_g , electron effective masses m_e^* , hole effective masses m_h^* , and electron (μ_e) and hole (μ_h) mobilities for $CsPbX_3$	129