Engineering Materials

Basudev Pradhan *Editor*

Perovskite Optoelectronic Devices

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Engineering Materials

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Dedicated to all researchers working in this field

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my parents, my wife Nupur, and daughter Oishi

Preface

Halide perovskites have emerged as an extremely promising material for a wide range of optoelectronic applications due to high absorption characteristics over a wide spectrum, long diffusion length, and bandgap tunability. For example, we have witnessed outstanding progress in perovskite solar cell research, from 3.8% efficiency to 26.1% in a single junction just over a decade after their inception and it is marching gallantly towards commercialization. Furthermore, perovskite-based optoelectronic devices consume a very low amount of energy. But with its advancing technology, it has been observed that these perovskite materials suffer from certain disadvantages related to device stability and also lead-based toxicity which are hindering the market commercialization of the perovskite-based devices. Hence, the research fraternity is trying to improve the device stability and is simultaneously looking for lead-free perovskite-based devices. The motivation behind writing this book arises from the belief that gaining a comprehensive understanding of the fundamental properties of perovskite materials and device designs can provide insights and solutions to these concerns. This book solely focuses on structural, electronic, and optoelectronics properties of perovskite materials followed by synthesis of bulk, two-dimensional (2D), and zero-dimensional (0D) perovskite materials and their uses in different optoelectronic device applications such as solar cells, light emitting diodes, photodetectors, neuromorphic devices, X-Ray detectors, lasers, etc. Particularly, this book would be helpful for scientists, technologists, and engineers who want to develop and optimize the perovskite-based optoelectronic devices toward product realization.

At first, this book reveals the detailed history of perovskite materials research and overview of materials properties, different device applications, and challenges. Chapters "Structural Properties of Perovskite" and "Electronic and Optical Properties of Perovskite Semiconductor" focus on understanding the structural, electronic, and optical properties of perovskite semiconductor and their origin. Chapter "Synthesis and Characterization of Perovskite Nanocrystals" of the book discusses about the synthesis of bulk, two-dimensional (2D), and zero-dimensional (0D) perovskite materials. Following the synthesis, it delivers the basic understanding and the optimization of the characteristics of these semiconducting materials using different characterization techniques. The theoretical insights of designing perovskite materials from basic understanding to advanced level by optimizing structural and optoelectronic properties for different optoelectronic applications are discussed thoroughly in the chapter "Theoretical Insights of Designing Perovskite Materials for Optoelectronic Applications." Later on, major emphasis is given on the different optoelectronic applications of the perovskite materials ranging from energy harvesting/generation, sensing, display to communication in the consecutive chapters. The halide perovskite materials are extensively studied from the beginning in the fabrication of low-cost high-efficiency solar cells with paramount success, so details research and development of perovskite solar cells from fundamental to large-scale commercialization in single junction to multijunction are discussed comprehensively in the chapters "Perovskite Solar Cells: Fundamental to Commercialization"–"Perovskite Based Tandem and Multijunction Photovoltaics." Despite of the unprecedented advancement of perovskite solar cells efficiency, large-scale commercialization has been hampered due to the degradation of the device under ambient operating conditions. In addition, these chapters also address concerns regarding stability as well as recent advancements to enhance the lifespan of perovskite solar cells. The exceptional performance of perovskite solar cells, boasting record-breaking efficiencies, has also propelled the exploration of these materials for various other optoelectronic applications such as light emitting diodes (chapter "Perovskite-Based Light-Emitting Diodes"), photodetectors (chapter "Perovskite Photodetector"), neuromorphic devices (chapter "Perovskite Based Neuromorphic Devices"), and X-Ray detectors (chapter "Perovskite X-ray Detectors"), lasers (chapter "Perovskite-Based Laser"). Chapter "Charge Transport Physics of Perovskite Field Effect Transistors"delivers more understanding on field-driven charge transport in perovskite semiconductor and various strategies employed to improve the device performance.

In recent times, research on two-dimensional perovskites also emerged to drive the legacy of the perovskite material which pushes the utility boundary of the materials from the limited applicability of three-dimensional perovskite. The chapter sixteen devotes the synthesis of "Two-Dimensional (2D) Perovskite and Its Applications". Lastly, one of the major obstacles for perovskite optoelectronic device commercialization is the "Instabilities and Degradation in Perovskite Materials and Devices" are discussed thoroughly in the chapter seventeen. The present book covers the complete fundamental knowledge about perovskite materials to perovskite optoelectronic device engineering with the future prospects. The different chapters of the book were contributed by the internationally acclaimed experts. I express my profound appreciation for their exceptional efforts to enhance the quality of this book. I hope this comprehensive book will ignite and inspire the readers to actively engage in the advancement of sustainable, affordable, and eco-friendly perovskite optoelectronic devices toward commercialization.

Ranchi, India Basudev Pradhan

About This book

Halide perovskites have garnered significant attention as an exceptionally promising material for a wide range of optoelectronic applications ranging from energy harvesting/generation, sensing, display to communication owing to their extraordinarily broad absorption characteristics, extended diffusion lengths, and bandgap tenability, etc. The book offers a concise overview of the most recent advancements and discussions in the perovskite semiconductors starting from the fundamental to different optoelectronic applications featuring contributions from leading scientists all over world in the field. This book solely focuses on structural, electronic, and optoelectronics properties of perovskite materials followed by synthesis of bulk, two-dimensional (2D), and zero-dimensional (0D) perovskite materials and their uses in different optoelectronic device applications such as solar cells, light emitting diodes, photodetectors, neuromorphic devices, X-Ray detectors, lasers, etc.

This book is suitable for graduate students, materials scientists, technologists, and engineers working in academia, R&D, and industry to develop and optimize the perovskite-based optoelectronic devices toward product realization.

Key Features

- Introduces the recent advances in the understanding of halide perovskites materials including relevant synthesis of bulk, 2D, and 0D materials.
- Theoretical insights of designing perovskite materials from basic understanding to advanced level for different optoelectronic applications.
- Basic understanding and the optimization of the characteristics of these semiconducting materials using different characterization techniques.
- Detailed advancement of perovskite solar cells from fundamental to large-scale commercialization in single junction to multijunction.
- Up-to-date explorations of perovskite materials for various optoelectronic applications such as light emitting diode, photodetectors, neuromorphic devices, and X-Ray detectors, and lasers.

- Covers field-driven charge transport properties in perovskite semiconductor.
- Discusses the synthesis of 2D perovskite and its different optoelectronic applications.
- In-depth examinations in instabilities and degradation in perovskite materials and devices.

Contents

About the Editor

Dr. Basudev Pradhan has been serving as an Assistant Professor in the Department of Energy Engineering, and Centre of Excellence (CoE) in Green and Efficient Energy Technology (GEET), Central University of Jharkhand, Ranchi, Jharkhand, India, since the year 2012. Before joining his present position, he was an "Alexander von Humboldt" research fellow at the University of Potsdam, Germany, for the period of 2010–2012. He was awarded the prestigious "Ramanujan Fellowship" from the Department of Science and Technology for the period 2013–2017 and was also awarded the SERC Fast Track research grant for Young Scientists. He was the recipient of the prestigious Bhaskara Advanced Solar Energy (BASE) Fellowship by the Indo-U.S. Science and Technology Forum (IUSSTF). Under this fellowship, he carried out research work on perovskite solar cells, phototransistors, and neuromorphic computers at the University of Central Florida in Orlando, Florida, USA, for the period 2018–2019. He was also awarded "Best Research Award-2023" in the School of Engineering and Technology, Central University of Jharkhand. He received his Master's degree in Physics from the University of Kalyani in 2002 and Ph.D. degree based on his thesis work on Organic Photovoltaics from the Jadavpur University and worked at the Indian Association for Cultivation of Science (IACS), India, in 2007. He spent more than three and half years as a postdoctoral fellow at Arizona State University, University of Wisconsin-Milwaukee, USA, and University of Surrey, UK, from 2007–2010. He is also working with Prof. Tsutomu Miyasaka, inventor of perovskite solar cell under India-Japan bilateral project. He is an experimental device physicist with expertise in the fields of next-generation solar cells and nano-electronic devices. He has published 46 research articles in different peer-reviewed journals with highlights in different international media and three US patents. He has also edited two books with reputed international publishers. He is one of the associate editors in *Applied Physics A: Materials Science and Processing*, Springer Nature Journal since 2014.

Introduction to Perovskite

Arup Mahapatra, Prashant Kumar, and Basudev Pradhan

Abstract Halide perovskite semiconductors have emerged as exceptionally promising materials for a plethora of optoelectronic device applications ranging from energy harvesting/generation to lighting and sensing due to their inherent chemical and photophysical properties. It took only just over a decade to reach the door of commercialization. The range of structural and compositional tunability has made its application more diverse and specific simultaneously. This chapter focuses on the evolution of perovskite material from its inception to its current applications, as well as discusses the exploration of its fundamental properties. The chapter also gives insights into understanding the material's physical and chemical properties and thus investigates avenues for implementation in different optoelectronic device applications. The chapter concludes with a discussion of the technological barriers towards commercialization, such as toxicity and stability.

Keywords Perovskite · History · Properties · Optoelectronic applications · Challenges

1 Background

Emerging perovskite-based semiconductor technology is exceptionally enriched with properties aligned in a way that contributes to the sustainable development of humankind. The trimmable and tunable characteristics of perovskite have projected it as a material of the future which is geared to sail through the challenges of the future. Future challenges of the world include challenges related to energy which has become the backbone of productivity and lifestyle. The increasing demand for energy is observed to go hand in hand with economic development all over the world.

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In order to fulfil the energy demands, avenues for more energy production have been explored with keeping energy sustainability in mind. Nevertheless, the majority of the energy requirement is satisfied through the utilization of non-renewable energy sources like coal and petroleum-based derivatives, which also creates challenges of various kinds of pollution. The critical issues of climate change and global warming have led to exploring the options of cleaner, greener and more sustainable forms of energy.

2 History of Perovskites

The journey of perovskites started in the year 1839 by Chief Mines Inspector of the Russian Empire August Alexander Kämmerer (1789–1858), who discovered a piece of chlorite-rich skarn in the Ural Mountains of Russia and sent the mineral to the German mineralogist and crystallographer Gustav Rose (1798–1873) for analysis. He studied its chemical composition and physical properties and also published a paper [1]. The mineral looks like a black in colour (Fig. 1a) and composition of calcium titanate ($CaTiO_3$). Kämmerer proposed that the newly discovered mineral be given the name as "Perovskite" in the honour of Russian politician (Minister of Internal Affairs under Nicholas I of Russia) and mineralogist Count Lev Aleksevich Perovski (1792–1856) [2, 3]. Figure 1b shows the chemically synthesized perovskite ($MAPbBr_3$) single crystal [4].

The crystal structure of Perovskites was studied by Victor Moritz Goldschmidt in the early 1920's and also first industrial patented by him (US1436164). He also proposed the fundamental rule related to distortion and stability of the perovskite structures known as tolerance factor explained in the later section [5]. Oxide forms of the perovskites was predominantly studied but its applicability as semiconductor was not suitable because of their large band gap (more than 2.5 eV). However, various other applications owing to the multiple properties such as piezoelectricity, ferromagnetism, high dielectric constant, ferroelectricity etc. have led to their use in different areas. Some of the materials of industrial importance are $BaTiO_3$ due to its high dielectric constant and application in superconductors, piezoelectricity etc. Other materials and their related properties such as piezoelectric properties of $Pb(Zr, Ti)O_3$, electro-strictive characteristics $Pb(Mg, Nb)O_3$, magneto-resistant $(La, Ca)MnO_3$, and multiferroic traits of $BiFeO_3$ have also found their way in various industrial applications [6–10].

In 1978, the exploration of organic–inorganic halide perovskite began by D. Weber at the University of Stuttgart in Germany, who conducted a study on the structure and properties of $MAZX_3$ (MA- Methyl Ammonium, Z-Pb/Sn, X-Cl/Br/I) [11]. After a span of nearly 16 years, specifically in 1994, scientists at the IBM T. J. Watson Research Center located in the United States accomplished the creation of light-emitting devices using luminescent perovskites comprised of organic–inorganic halides. Furthermore, in 1999, a group led by M. Chikao at the AIST situated in Tokyo, Japan, published a report documenting the utilization of rare-earth-based

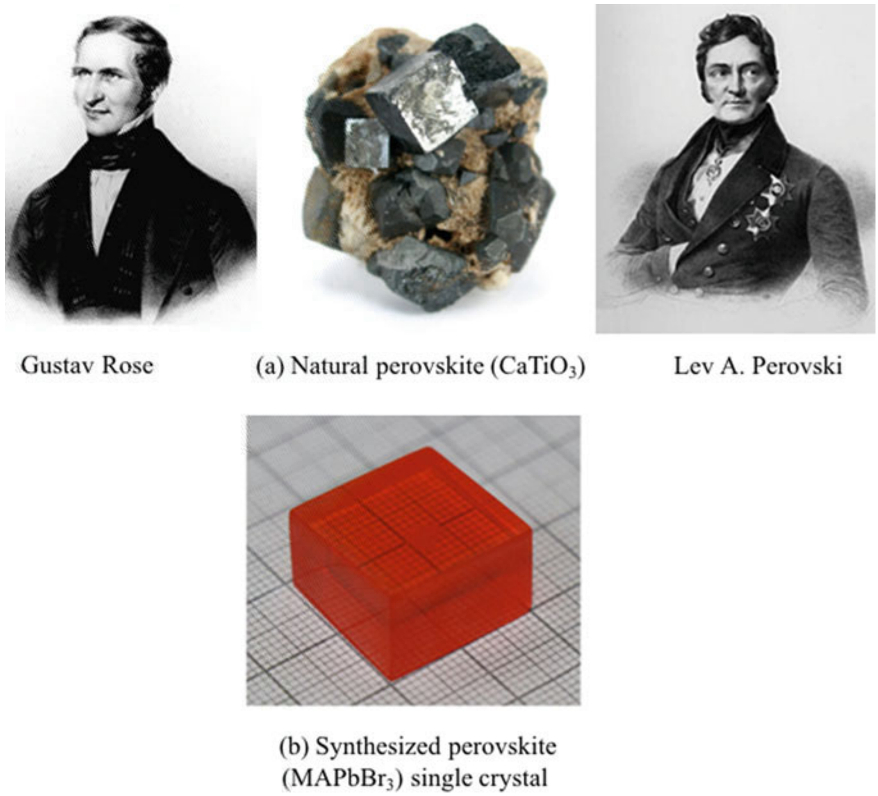

Fig. 1 **a** Natural mineral perovskite ($CaTiO_3$) (image taken from copyright © Rob Lavinsky, iRocks.com—CC-BY-SA-3.0). **b** Synthesized perovskite ($MAPbBr_3$) single crystal Reprinted with permission from Ref. [4]. © copyright 2020, American Chemical Society

perovskite compounds as an optical absorption layer within solar cells. A variety of compositions of smaller and large cations and their properties were studied by Mitzi towards the ending of the last century.

However, the application of perovskite material in photovoltaic began recently in the year 2005 [12]. The perovskite material was used as sensitizer molecule absorbed by the Titanium dioxide (TiO_2) for the first time in Dye-sensitized solar cell (DSSC) and a efficiency of 3.81% was achieved. This was the first study reported in the area of perovskite application in PV domain by Prof. Tsutomu Miyasaka and his team at The University of Tokyo, Tokyo Polytechnic University, and Toin University of Yokohama, Japan in the year 2009 [12, 13]. Later on the need for development of solid-state solar cells due to the disadvantages posed by liquid electrolyte in DSSCs led to the development of solar cells with perovskite material as an absorber material. One of the breakthrough results demonstrated by Tsutomu Miyasaka and Henry Snaith group by the application of Al_2O_3 scaffold layer for mixed halide perovskite ($CH_3NH_3PbI_{3-x}Cl_x$) with Spiro-OMeTAD(2,2′,7,7′-tetrakis(N,N-dimethoxyphenylamine)-9,9′-spirobifluorene) as the hole transport layer(HTL) in a solid-state perovskite cells. This study pushed the efficiency of perovskite solar cells to 10.9% and attracted the global scientific community for advancements in this field [14]. Onward the perovskite solar cell makes several milestones and reaching to power conversion efficiency of 26.1% which is very close to conventional silicon solar cells and 33.9% efficiency in perovskite/silicon tandem solar cell [15]. The ground-breaking innovation triggers scientific community to investigate the inherent optoelectronic characteristics of perovskites. The metal halide perovskite emerges as major semiconducting martials due to high absorption characteristics over a wide spectrum, long diffusion lengths, and bandgap tunability for different optoelectronics. In the 2014, Xing et al. demonstrated high optical gain and stable amplified spontaneous emission using solution-processed organic–inorganic halide perovskites with a threshold of 10 ± 2 J cm^2 at room temperature [16], and the first laser work in this field without an external cavity was reported by Zhang et al. in the same year [17] and Deschler et al. reported optically pumped vertical cavity laser using a layer of mixed halide perovskite materials [18]. Around the same year, Friend and colleagues reported the creation of the first room-temperature perovskite light emitting diode with an external quantum efficiency (EQE) of 0.76% using halide perovskite materials [19]. The next level of development happen in the perovskite optoelectronic devices by tuning the dimensionality of the perovskite martials from bulk 3D structure to layer and perovskite nanocrystal due to enhanced photoluminescence (PL) and electroluminescence (EL) efficiency in comparison to its bulk counterpart [20, 21]. In 2018, Chen et al. demonstrated X-ray detector using all inorganic $CsPbBr_3$ Nanocrystals with a low detection limit of 13 nanograys per second [22]. The summary of major discoveries and breakthroughs in perovskite optoelectronics research are shown in the Table 1. Meanwhile, the main challenges like device stability is also showing improvement day by day. Therefore, perovskite materials have emerged as an important semiconductors, finding applications not only in solar cells, LEDs, photodetectors, and lasers, but also in facilitating the integration of diverse optoelectronic devices. This progress has injected fresh momentum into the commercialization of optoelectronic devices.

3 Perovskite Structure and Its Properties

Typically, a perovskite structure refers to inorganic metal oxide with a crystal structure of ABO_3 such as $BaTiO_3$, $CsTiO_3$, $SrTiO_3$ etc. and after oxygen is replaced by halogens, halide perovskite with crystal structure ABX_3 is formed. Halide perovskite has captured a great deal of attention and has been under investigation to get a deeper understanding of perovskite physics and explore its optical, electrical, and magnetic applicability. The ideal structure of the perovskite is uncommon, which is a cubic structure comprising the BX_3 octahedra at the corner of the cubic structure and amidst the octahedra lies A site cation is given in Fig. 2 where B has a sixfold coordination

Table 1 Summary of major discoveries and breakthroughs in perovskite research

Year	Short description	Lead researcher(s)
1839	The identification and designation of perovskite ($CaTiO_3$) occurred when a sample from the Ural Mountains in Russia, specifically from a skarn known as "Schisto chloritico," was examined	Gustav Rose (Prussia)
1851	First documented synthesis of $CaTiO_3$ (flux growth)	Jacques-Joseph Ebelmen (France)
1898	The initial synthesis of a compound with a perovskite structure occurred with the creation of $NaNbO_3$	Per Johan Holmquist (Sweden)
1912	The orthorhombic symmetry of $CaTiO_3$ was verified through confirmation experiments	Ove Balthasar Bøggild (Denmark)
1922	The first patent in the industrial sector was filed for the production of a $CaTiO_3$ pigment	Victor M. Goldschmidt (Norway)
1925	First description of the perovskite crystal structure	Thomas F. W. Barth (Norway)
1940	The discovery of ferroelectric ceramics possessing a high dielectric constant was made with the identification of $BaTiO_3$	B. M. Vul and I. M. Gol'dman (Physical Institute, USSR); A. von Hippel and co-workers (MIT, USA)
1949	The pioneering observation of the first "layered perovskites," known as Aurivillius phases, commenced with the discovery of $Ca_2Nb_2Bi_2O_9$	Bengt Aurivillius (Stockholm University, Sweden)
1950	The discovery of ferromagnetism and magnetoresistance in mixed-valence manganites, specifically $La_{1-x}(Ca, Sr, Ba)_xMnO_3$, marked a significant breakthrough	G. H. Jonker and J. H. Van Santen (Philips, Netherlands)
1952	The initial investigation into catalysis within perovskite materials was conducted	Giuseppe Parravano (Princeton, USA)
1955	The magnetic structures of $La_{1-x}Ca_xMnO_3$ perovskite compounds were examined and analyzed	E. O. Wollan and W. C. Koehler (Oak Ridge National Lab, USA); J. B. Goodenough (MIT, USA)
1955	Development of Pb zirconate–titanate piezoelectric materials (PZT, $PbZrxTi1 - xO_3$)	Bernard Jaffe (Clevite Corp., USA) and co-workers
1958	Starting with $Sr_3Ti_2O_7$, the process involves creating layered derivative (Ruddlesden-Popper) phases	S. N. Ruddlesden and P. Popper (British Ceramic Research Association, England)
1964	Developed perovskite-based solid electrolytes for fuel cells	Compagni e Generale d'Electricité (Paris, France)
1970	Discovery of perovskite-type cobaltite catalysts	D. B. Meadowcroft (CERL, England)
1971	Creation of cathode catalysts based on perovskite for electrochemical cells that can efficiently convert alcohols into ketones	Exxon Research Engineering (Linden, N. J.)

(continued)

Table 1 (continued)

Year	Short description	Lead researcher(s)
1975	Superconductivity has been observed in perovskite materials, specifically in compounds such as $BaPb_{1-x}Bi_xO_3$	Arthur W. Sleight (Dupont, USA) and co-workers
1975	Manufactured the first gas sensors based on oxide perovskites	Hitachi (Tokyo, Japan)
1978	The initial creation of organic–inorganic halide perovskites	D. Weber/University of Stuttgart (Stuttgart, Germany)
1981	Proton conduction in anion-deficient cerate perovskite materials ($SrCe1-xREExO_3$ -α) was identified	Hiroyasu Iwahara (Tottori University, Japan) and co-workers
1981	The implementation of lasers utilizing perovskite crystals was introduced	GTE Laboratories (Waltham, Mass
1986	The identification of high-temperature superconductivity in ceramics composed of perovskite-type cuprates was made	J. G. Bednorz and K. A. Müller (IBM, Switzerland)
1994	The advancement of hybrid organic–inorganic halide perovskites for the application of thin-film transistors was achieved	David B. Mitzi (IBM, USA) and co-workers
1996	Cesium-germanium halide salts with a perovskite structure were introduced for applications in optoelectronics	Boeing North America (Seal Beach, Calif.)
1999	A solar cell's optical absorption layer was developed by utilizing a perovskite crystal structure rare earth oxide	Murase Chikao et al./National Institute of Advanced Industrial Science and Technology (Tokyo, Japan)
2009	Perovskite-sensitized solar cell was created with 3.81% efficiency, utilizing the hybrid halide perovskite $CH_3NH_3PbI_3$	A. Kojima, T. Miyasaka et al. (Tokyo Polytech, The University of Tokyo, and Toin University of Yokohama, Japan)
2014	Preparation of first perovskite ($MAPbBr_3$) nanocrystal	Pérez-Prieto and colleagues (Universidad de Valencia, Valencia, Spain)
2014	Perovskite light emitting diode	Richard H. Friend, and colleagues (Cavendish Laboratory, University of Cambridge, UK)
2014	Perovskite based laser	Tze Chien Sum, Nripan Mathews and coworker, Nanyang Technological University, Singapore
2018	$CsPbBr_3$ NCs X-ray detector	Xiaogang Liu and co-worker, National University of Singapore, Singapore, Singapore

(continued)

Table 1 (continued)

Year	Short description	Lead researcher(s)
2021	25.8% efficiency in single junction solar cells	Sang Il Seok and co-worker, Ulsan National Institute of Science and Technology, South Korea
2023	26.1% efficiency in lead halide based perovskite solar cells in single junction	Northwestern University and the University of Toronto, and also University of Science and Technology of China
2023	33.9% efficiency for perovskite/silicon tandem solar cell	LONGi Green Energy Technology Co. Ltd, China

number, X has two-fold, and A has 12 fold cuboctahedra coordination number. The unit cell of the perovskite comprises of A cation at the corners and B at the body-centered position and X placed at the face-centered position as shown in Fig. 2 [24]. The crystal structure possesses the Pm-3m space group with a Z value of 1. The A atoms occupy the Wyckoff position 1b at coordinates ½, ½, ½. The B atoms reside in position 1a at coordinates 0, 0, 0. Additionally, the X atoms are located at the special positions 3d with coordinates ½, 0, 0; 0, ½, 0; and 0, 0, ½ [25]. In a crystal structure ABO_3 the charge stability is as $q^A + q^B + q^O = 0$, which means $q^A + q^B = - q^O = 6$. And for halide perovskite ABX_3 crystal structure $q^A + q^B + q^X = 0$ which gives $q^A + q^B = - q^X = 3$. The possible combination of A-B oxidation states could be I-V-O_3, II-IV-O_3, III-III-O_3 for ABO_3 structure and I-II-X_3 only for ABX_3 structure. The I-II-X_3 combination in ABX_3 can be of inorganic–inorganic and organic–inorganic depending upon the structural limitation which the perovskite crystal possesses.

In the ideal case, the distance between B-X should be a/2 where a is the cell length and the distance of A–X should be $\sqrt{2}a/2$. Also, in case of ideal structure it holds the relation given by the sum of ionic radii of A (R_A) and X (R_X) i.e., $R_A + R_X = \sqrt{2}(R_B + R_X)$ [26]. However, it was observed that this perovskite structure is retained even

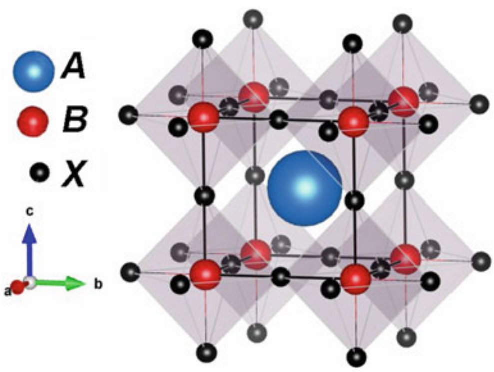

Fig. 2 Unit cubic cell of ABX_3 structure Reproduced with permission [23]. © 2016, The Royal Society of Chemistry

if these relations do not hold and defined it with a tolerance factor, 't' also known as Goldschmidt tolerance factor given by:

$$t = \frac{R_A + R_X}{\sqrt{2}(R_B + R_X)} \tag{1}$$

For ideal perovskite the value of t is 1, however perovskite structure is also found for the lower values than 1; t ($\sim 0.75 < t \leq 1$) [27]. In these types of cases, the structure is found to be slightly distorted into structures such as rhombohedral, orthorhombic or tetragonal symmetries. These distortions also give birth to interesting magnetic and electronic properties. The structural stability of the perovskite is guided by the sizes of the cations and anions and their geometrical structure. On balance, the size of the A site cation is limited by the vacancy space. A site cation size greater than vacancy would be led to tailoring the perovskite by increasing the gaps between the octahedra in the direction thus lowering the dimensionality. In general, metal halide perovskite, organic cation size to be smaller to fit into the corner-sharing metal halide octahedral, but if it is bigger, in that case perovskite 3D structure framework need to be change intro different 2D, 1D, or 0D dimensionality as shown in the Fig. 3. BX_6 octahedral are isolated in the case of 0D structure, whereas BX_6 octahedra are connected in a chain at the edges or faces and corners for 1D structure. In the case of 2D structure, BX_6 octahedra are connected in the layer in the corners. But in the all cases, organic cations are surrounded by the inorganic framework. In the present era, nanostructure perovskite provides wide range of opportunities for engineering diverse materials compositions for specific application such as solar cell, photodetector, light emitting diodes, etc. by simple synthesis methods.

The most common organic- inorganic combination in I-II-X_3 structure is $MAPbI_3$ and $FAPbI_3$ which has been studied to uncover the perovskite physics. The crystal structure of $MAPbI_3$ exhibits temperature-dependent variations and can transition between orthorhombic, tetragonal, and cubic Bravais lattices as the temperature increases. This can be distinguished from the Braggs' peak position and intensities through X-ray diffraction but is not clearly distinguishable from the TEM image [29]. The phase of the $MAPbI_3$ crystal is orthorhombic at T < 165 K, is tetragonal from 165 to 327 K and is cubic when the T > 327 K. The stability at different temperature can be confirmed from the difference in enthalpy which is around 2 eV in case of the most stable phase tetragonal and is 90 eV in case of the cubic phase [30–32]. Typically for perovskite materials, the absorption coefficient is very high up to 10^5 cm^{-1} over a wide spectrum, which originates due the direct band gap nature of the materials as well as the loan pair electron transition between valance band and conduction band. The absorption and the carrier diffusion lengths, diffusion constants and life times can be tuned by varying the concentration of the halides in the perovskite. Such observations can be confirmed by the PL spectra. The long bandwidth of compositional variation and resulting interplay of the structures gives rise to a variety of properties which has been utilized for various applications.

The underlying physics and chemistry of perovskite materials make it a quintessential material for a huge range of evolving applications. Various ab- initio

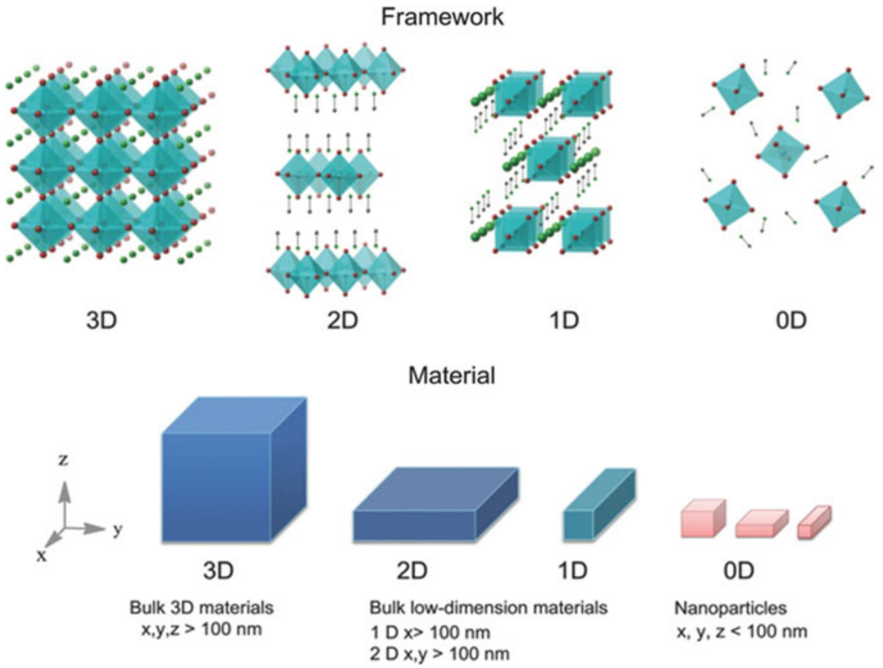

Fig. 3 Schematic illustration of perovskite framework with different dimensionalities (3D, 2D, 1D, and 0D) and (bottom) materials with different degree of confinement. Reprinted with permission from Ref. [28]. © Copyright 2015, WILEY–VCH

calculation for $MAPbI_3$ based on Density Functional Theory (DFT) and Many-body Perturbation theory (MBPT) has been performed to estimate the carrier density of the material. Band structure and density of states have also been predicted. The calculations have revealed remarkably high mobilities for both lead-based and tin-based perovskites. For lead-based perovskite, the electron mobility (μ_e) ranges from 3100 to 1500 cm^2/Vs, while the hole mobility (μ_h) ranges from 500 to 800 cm^2/Vs at a charge carrier concentration of approximately 10^{16} cm^{-3}. As for tin-based perovskites, the electron mobility (μ_e) ranges from 2700 to 1300 cm^2/Vs, and the hole mobility (μ_h) ranges from 3100 to 1400 cm^2/Vs under similar charge carrier concentrations.

In experimental observations, the carrier mobility values of perovskite materials typically fall within the range of tens of cm^2/Vs. Such high mobilities can be attributed to the low effective mass of electrons and holes, which are 0.2333 m_0 and 0.2583 m_0 (where m_0 represents the rest mass of the electron) in the case of lead-based perovskite, and 0.3040 m_0 and 0.1906 m_0 for tin-based perovskite, as confirmed by magneto-absorption measurements. Another contributing factor to these high mobilities is the weaker carrier-phonon interaction, which is supported by the large diffusion length observed in previous experiments [33–35]. A diffusion length exceeding 1 μm has been observed, making the material highly suitable for optoelectronic applications [33]. Additionally, halide perovskites display ambipolar charge transport properties due to the low effective mass of electrons and holes, which play a vital role in carrier transport in perovskite-based devices.

In a study focusing on $CH_3NH_3PbI_3$ perovskite, the defect properties were investigated, revealing that shallow levels are created by point defects with low formation energies, while deep levels are formed by point defects with high formation energies. Notably, intrinsic surfaces and grain boundaries do not generate deep levels. This phenomenon can be attributed to the weak antibonding coupling between the lone pair of s orbitals in Pb and p orbitals in X, thus it exhibiting high iconicity due to this energy state are remain unaffected by the external defect levels, which lead to high defect-tolerance and the large lattice constant of the perovskite. Usually, the electronic and ionic molecular dielectric response is found in solid-state dielectric, but an additional molecular dielectric response arises in materials with the permanent dipole. The orientation effect arising here has a temperature and frequency-dependent response [36].

Undeniably, the convergence of such characteristics and features makes it a semiconductor of demand in the present era. This material offers opportunities to directly adjust bandgaps by altering compositions, featuring strong light absorption, balanced electron/hole effective masses, and resistance to defects when high-quality crystals and films are developed. Moreover, it provides avenues to independently and synergistically modify structural, optical, and electronic properties, positioning it as a versatile semiconductor for diverse applications [37]. This advancement has facilitated easy correlation between structure and property, allowing for rapid prototyping and optimization of devices. As a result, perovskite is being promoted as a superior semiconductor for a wide range of accessible optoelectronic applications in future generations.

4 Perovskite Optoelectronic Applications

Metal halide perovskite has emerged as a significant material with distinctive photophysical properties, finding applications in diverse fields of optoelectronic devices, including energy harvesting/generation, sensing, display, and communication, as illustrated in Fig. 4. Particularly in light-harvesting devices like solar cells and photodetectors, these materials absorb photon energy and convert it into charge carriers. Notably, the efficiency of perovskite solar cells (PSCs) has experienced remarkable advancements within the past two decades since their inception [15]. The reasons behind this progress are owed to the special properties which these materials have shown such as high absorption over a wide spectrum, longer diffusion lengths and defect tolerance.

The PSCs came into existence as extended research on DSSCs which are basically a thin film solar cell. This PV material mostly uses metal halide perovskites sandwiched between two transport layers and electrodes. The compositional perovskite material is generally known for its metal oxides and chalcogenides. The halide ions

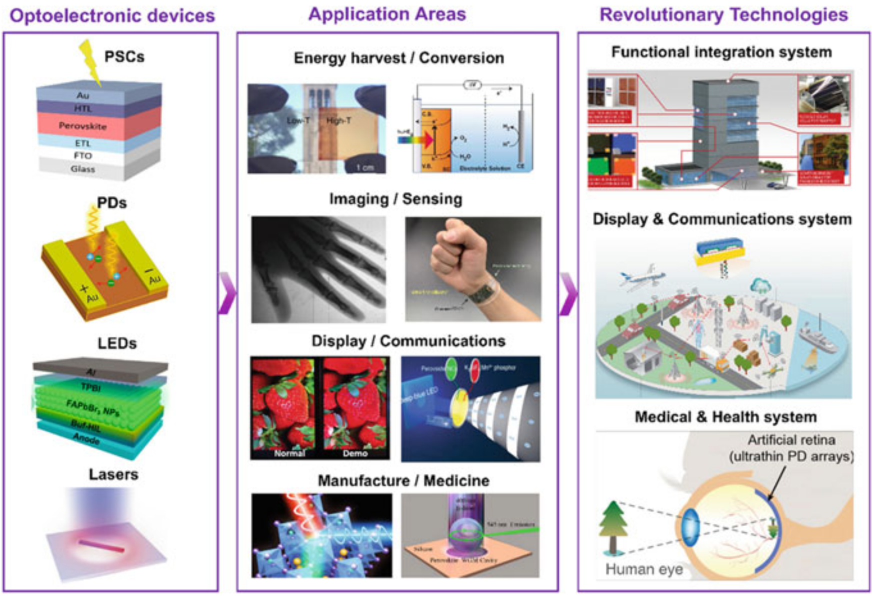

Fig. 4 Schematic diagram to represent different optoelectronics application of perovskite materials ABX_3 with functional integrations (acronyms: PSCs perovskite solar cells, LEDs: light-emitting diodes, PDs: photodetectors) Reproduced under CC BY license [38] © 2023 Springer nature

in the metal perovskites also provides superior ionic properties due to its electronegativity making them suitable as ionic conductors. To comprehend the property variations and the influence of substitutions at different positions in perovskite materials, a thorough understanding of their structure is essential. Such knowledge of structural and compositional variations plays a pivotal role in identifying materials suitable for photovoltaic (PV) applications. The exceptional performance of perovskite solar cells, boasting record-breaking efficiencies, has also propelled the exploration of these materials for various other optoelectronic applications. Perovskite based photodetector is a type of sensor which converts light into electrical signals is of great use in optical communication and monitoring. The excellent photophysical characteristics such as band-gap tuning, carrier mobility and long diffusion lengths have paved the way for there application as photodetectors [39]. The photodetectors can be made into vertical or lateral configurations. The vertical configurations of photodetectors are similar to perovskite solar cells structure. On the other hand, lateral configuration of photodetector such as phototransistor, and photoconductor, are made of metal-perovskite-metal and the field effect transistor configuration for light detection and also signal amplification simultaneously. So far the perovskite based photodetectors are able to detect light intensities rang from 1 pW/cm^2 to ~ 100 mW/cm^2. Various compositions of perovskite materials have been found to sense a broad spectrum of UV to almost X and γ-rays [40].

The technological potential of the perovskites is also broadened to other sustainable electronic devices for artificial intelligence platform as synaptic and memory application. Resistive Random Access memory devices (ReRAM) are high memory density, next generation non-volatile memory devices with low power consumption and fabrication cost [41]. Generally, ReRAM is a two terminals metal–insulator-metal stack where the top electrode is externally biased and the bottom electrode is grounded. The electrochemical metallization mechanism defined as conductive filament formation by the active electrode and valence change mechanism defined as electronic conductivity change due to enriched oxygen with the inert oxygen occurs during the high resistance state to low resistance state switching and vice versa in the devices. The functioning of the device is related to the soft break down of the insulator material in between thus leading to changes in resistances by applying the external biases [42–44]. Considerable tuning of the A, B, X in the ABX_3 perovskite structure effects bandgap leading to various Schottky barrier heights. This helps us to know the resistive ON/OFF resistive ratio in the memory devices [45, 46]. On the other hand, perovskite material based synaptic and neuromorphic devices, has taken lot of research interest for various applications which need to meet the urge of Internet of Things (IoT) and Big Data. Inorganic perovskite based ReRAM devices with $CsPbI_3$, or $CsPbBr_3$ as active material, have been investigated to overcome the bottleneck of thermal and photo instability, storage, fabrication, operation in ambient condition for the future application [13, 47–49]. In the case of perovskite based light emitting devices, charge carrier injected both side of electrode of perovskite layer in a sandwich structure under forward bias, where the injected charge carrier recombines and emit light. Perovskite light emitting diode can now able to emit ~ 1 $\mu W/cm^2$ to ~ 10 mW/cm^2 with different colors. Whereas in case of perovskite based laser, light amplification is done by optically pumping and with the carrier density reaches over ~ 10^{18} cm^{-3}.

Such intriguing properties has added more doors to the room of applicability of perovskite materials. It has been widely investigated for its application in energy harvest, storage, imaging, sensing, communication, manufacture, medicine as depicted in the Fig. 4. In the development of electronic communication system, the perovskites can also contribute a lot in the field of consumer based electronics including mobile communications, advanced computing technologies, PeLEDs, photodetectors, digital camera, smart phone and medical devices components [50]. With the rise in automation, it can also use in automated driving, environmental monitoring, optical communication, and biosensors. Apart from this its use can be expanded to modernising the laser technology. In health and medical system, it includes application in biomedicine, imaging and monitoring techniques. This also requires flat detectors for diagnostic and interventional applications like in X-ray imaging procedure for which perovskites are good candidate [51]. Recently, $CsPbBr_3$ perovskite quantum dots were utilised in the application of X-ray scintillator for the facile visualization of the X-ray radiography [22]. Its exhibition of chiroptoelectronic property along with circularly polarized photodetector has made it more promising for flexible integrated devices application [52]. In sum, the functional integration system based on perovskite optoelectric devices may become one of the most disruptive technologies for the smart cities, IoT, and wearable electronics. However, there are few issue like instability and toxicity in general with the perovskite optoelectronic device, which hinders the large scale commercialization. Further, due to extraordinary success in perovskite optoelectronic device by taking the advantages of low cost, semi transparency, flexibility, etc. now the scientist/researcher has been exploring for smart integration of devices including smart photovoltaic devices, self-powered system, smart display, wearable electronics, health care devices, etc. The variety of applications has been briefly discussed in the later sections of the book.

5 Challenges and Perspectives of Perovskite

As mentioned previously, perovskites have a broad range of applications. However, their utility has been limited by two main factors: (1) toxicity associated with lead-based perovskites and (2) stability concerns. To understand the instability of perovskites, we need to recognize that it is inherent in their geometric structure. Perovskites degrade when exposed to external factors such as moisture, oxygen, heat, light, external bias, and contact layers. These factors alter the stabilized range tolerance factor, which is ideally favored to be between 0.8 and 1 for achieving an ideal cubic shape [53]. Moreover, the stability is also related to the ion migration in the perovskite material which also gives rise to the hysteresis, phase segregation, slow photoconductivity response, and device performance degradation in the devices. Other sources of instability could be stress in the film due to polycrystallinity, smaller grain sizes [54, 55]. However, this issue can be addressed by incorporating additives, which not only enhance the stability of the material but also improve its optical parameters for enhanced performance. Cation engineering and solvent engineering have also contributed to the improvement of device performance and stability. By introducing additives, defects can be passivated, addressing the root cause of instability. Furthermore, the fabrication of various all-inorganic devices has demonstrated increased stability, as organic molecules within the perovskite are more susceptible to instability when exposed to external factors. Moreover, dimension tailoring of the compound has shown promising results in enhancing the stability of perovskite devices. It has been observed that 2D perovskites are more efficient and stable compared to their three-dimensional counterparts. On the other hand, suitable encapsulation techniques also provide a stable solution for mitigating instability concerns.

Another significant challenge in perovskite-based devices is the toxicity associated with lead, a predominant component. Lead has been identified by the World Health Organization (WHO) as one of the top ten toxic elements globally, posing severe hazards to human health. Its contamination can give rise to various illnesses affecting the nervous system and the reproductive system. Once released into the environment, lead can easily dissolve in water, leading to contamination of aquifers and an increase in lead concentration beyond normal levels in the ecosystem. In perovskite devices, lead exists in an ionic form, amplifying its potential dangers for commercial applications.

However, this concern can be effectively addressed through cation substitution methods, replacing lead with group 14 elements. Alternatively, a mixture of group 15 and 13 cations can be used to form double perovskites as a replacement. Another crucial approach involves the development of well-equipped encapsulation techniques for perovskite devices. Additionally, efforts have also been made to develop lead-free perovskites with comparable efficiency and stability, which will be discussed in later sections of this book.

6 Conclusions

In conclusion, perovskites have a multitude of benefits and have therefore arisen as a miraculous material with a varied range of utility. During its brief voyage, this material has demonstrated encouraging performance in comparison to other materials; however, it has encountered several constraints. With its practicality, the resolution of these challenges will spark a revolution in the modern world. Nevertheless, further potential exists for enhancing its overall performance, thereby facilitating the advancement of optoelectronic technology to the next level.

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