Study on the two-dimensional perovskite materials and three-dimensional perovskite materials

Haiyue Yu [1, *]

[1]Department of microelectronics, Dalian University of Technology, Dalian, China

*Corresponding author: 1282707574@mail.dlut.edu.cn

Abstract: Perovskite composite oxides exhibit special crystal structures. Their properties could be enhanced greatly after doping. Many disciplines, including physics, chemistry, and materials, are focusing their researches on perovskite materials. In standard perovskite structures, oxides with different composites can be created by substituting A or B site ions with other elements. In this way, a brand-new class of functional materials with tunable properties, outstanding functionality and broad applications can be designed. Perovskite is a kind of material with various excellent properties. It can be divided into two dimensional (2D) perovskite materials and three dimensional (3D) perovskite materials. Both of them have many outstanding advantages. However, the structures of the two kinds of materials are quite different. As a result, their performances, characteristics and applications are quite different as well. These similarities and differences are worth studying. Therefore, in this work, the properties and applications of 2D and 3D perovskite materials are investigated.

Keywords: Perovskite, two-dimensional material, three-dimensional material.

1. Introduction

Perovskite is referred to a class of ceramic oxides with the structural formula ABO_3. $CaTiO_3$ chemical that was the original form of this type of oxide was found in perovskite, and thus the name. Due to the various properties of such compounds, they are extensively utilized and been researched. It is often referred to as the "113 structure" by physicists and chemists because of their ratio of the elements in their chemical formula (1:1:3). It takes the shape of a cube. The phase transition from the high temperature variation to the low temperature form often results in a stripe of parallel crystal edges on the cuboidal crystal. As an innovative inorganic nonmetallic material, the perovskite-type composite oxide ABO_3, has special chemical and physical properties [1]. Typically, an rare earth or alkaline earth element ion occupies the A site, whereas a transition element ion occupies the B site. Other metal ions with a similar radius can replace the A or B site, effectively maintaining the crystal structure. It also provides a perfect sample for theoretical research on the surface and catalytic characteristics of catalysts. Perovskite materials, which are a new category of functional materials, have potential applications in the area of environmental protection due to their good electromagnetic properties, stable crystal structure, electrocatalysis, and so on.

Usually, bulk materials are commonly used, which is the so-called three dimensional (3D) materials. However, compared with 3D materials, two dimensional (2D) perovskite materials have superior performance in some aspects [2]. Two-dimensional (2D) materials refer to materials with only one dimension of nanometer size, including planar films, nanocoatings and superlattices with only atomic thickness. They are one of the hot research topics in the field of materials today. Compared with bulk materials, 2D materials have many unique properties, which can be mainly summarized as the following four points :(1) they have a large plane, ultra-thin thickness and large specific surface area, which can be used as a catalytic material carrier; (2) The activity space of electrons is confined to an isolated plane and has a series of new electronic properties; (3) There are strong chemical bonds in the layers and van der Waals forces between the layers, so the mechanical properties of the material are better; (4) The material has strong flexibility and light transmittance, which can be used as photoelectric devices with high elasticity and high transparency. As the star unit of 2D materials, graphene was considered to be the hotspot since it was discovered. In addition, with the rapid development of 2D materials, more and more new 2D materials have been reported, such

as hexagonal boron nitide (h-BN), transition-metal dichalcogenides (TMDs), black phosphorus, etc. While new materials continue to emerge, the applications of 2D materials in microelectronics, catalysis, energy storage, photoelectric conversion and other fields are gradually expanded. Till now, the research of 2D and 3D perovskite materials are still lacking. Therefore, in this work, the differences between 2D perovskite materials and 3D perovskite materials would be summarized, as well as the characteristics and applications of these two materials.

2. The structure and development of organic-inorganic 2D perovskite materials

Organic-inorganic hybrid perovskites have ABX_3 structure, where A is generally $CH_3NH_3^+$, $HC(NH)_2^+$; B is generally Pb^{2+}, Sn^{2+}; X is usually Br^-, Cl^-, I^-. If $CH_3NH_3^+$ is replaced with Cs^+, the inorganic halide perovskite material can be obtained.

The stacking mode of the regular octahedral $[BX_6]^{4-}$ structure of perovskite determines that the 2D morphology of perovskite materials is easy to be achieved. As early as 2014, Qihua Xiong's research group from Nanyang Technological University in Singapore carried out research on the preparation of 2D perovskite nanomaterials by vapor deposition [3]. In subsequent work, Haizheng Zhong's research group of Beijing Institute of Technology discovered the luminescence induced by the aggregation of 2D layered perovskite materials in mixed solvents, and prepared 2D layered perovskite nanomaterials by reprecipitation method [4]. However, in these studies, the number of perovskite layers is more than 20 layers, which does not show the expected quantum confinement domain effect. Later, William A. Tisdale's research group from Massachusetts Institute of Technology paid attention to the existence of perovskite nanosheets during the synthesis of perovskite nanocrystals [5]. They proposed the idea of controlling the number of perovskite layers by using long alkyl chain amine salts, and they investigated how the number of layers affected the optical characteristics of perovskite nanosheets.

Subsequently, Alexander S. Urban's research group at the University of Munich, Germany also prepared $L_2[MAPbBr_3]_{n-1}PbBr_4$ nanosheets with controllable layer number by introducing the strategy of long alkyl chains [6]. At the same time, Peidong Yang's group at the University of California, Berkeley prepared $(C_4H_9NH_3)_2PbBr_4$ nanosheets with tunable layers of 3 ~ 22 by a method of controlling crystal growth on silicon-based substrates [7]. The development of these studies has aroused great interest in 2D perovskite materials and attracted many researchers to carry out research in the field of 2D perovskite nanomaterials.

The chemical composition of 2D halide perovskite material is A_2BX_4 and its crystal structure is shown in Figure. 1(a). Lattice distortion caused by the tilt and rotation of an octahedron, a cell containing two halide octahedrons connected by angular sharing $[BX_6]^{4-}$, the monovalent cation A^+ with a certain size is located in the center, which plays the role of supporting the 2D grid structure. The calculation results of determining whether Rb_2SnX_4 and other structures are stable based on tolerance factor T are shown in Figure. 1(b). Negative formation energy represents the energy required to be absorbed when halide perovskite decomposes binary halides [1]. According to the principle of minimum energy, structures with negative formation energy are more stable. Taking t=0.81 as the dividing line, the system with t<0.81 has a positive formation energy, and the system with t>0.81 has a negative formation energy [1].

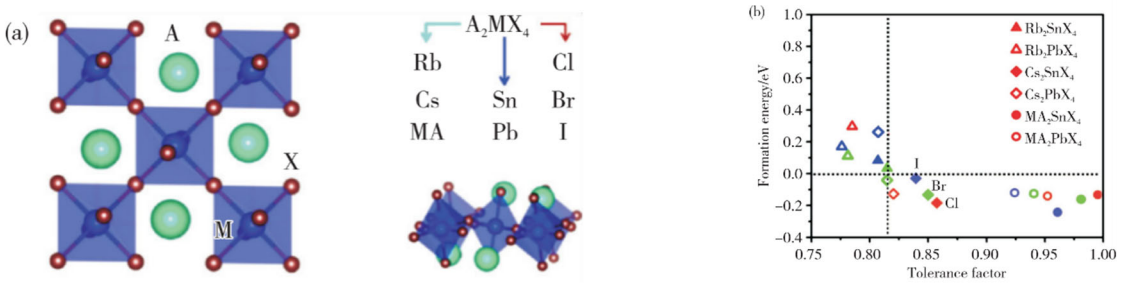


Figure 1.(a)The schematic diagram of 2D halide perovskite structure and its optional components ; (b) the system under consideration forms the energy distribution and tolerance factor, The vertical dashed line represents t=0.81 and the horizontal dashed line is zero formation energy [1]

Rp-phase perovskite was obtained after the bulk structure was dimensionality reduced by organic cations, and its general formula can be expressed as $A_{n-1}A_2'B_nX_{3n+1}$. The size of the two cations, or ionic groups, A and A' can be changed to alter the interlayer spacing of perovskite [8]. The parameter n indicates how many metal halide monolayer sheets are sandwiched between the insulating layers. When n decreases, the quantum confinement effect increases. Two extreme cases are considered. When $n = \infty$, the general formula of the material is ABX_3, which exhibits a long-range ordered cubic structure and is a 3D bulk perovskite material. When n = 1, the general expression of the material is A_2BX_4, which is represented as A 2D layered perovskite with skeleton atom B supporting two A atoms (or atomic groups) [9]. As shown in Figure. 2, the RP phase perovskite has a "sandwich" structure, and the general formula of its changing process with the change of the layer number n is $A_{n+1}B_nX_{3n+1} = (ABX_3)_n + AX$ [10].

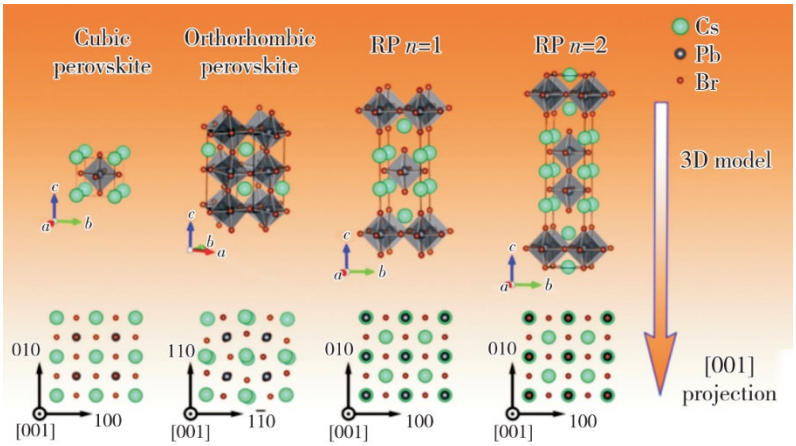


Figure 2. The crystal structure diagram of conventional $CsPbBr_3$, perovskite phase (cubic and orthogonal) and Rp-phase (n=1,2) [11]

Compared with 3D halide perovskite, 2D halide perovskite has better stability, which is specifically manifested in the higher energy of 3D halide perovskite system than 2D halide perovskite [12]. Yang et al. believe that this phenomenon is due to the higher tolerance of deformation in 2D materials, which increases the Coulomb interaction and improves the stability of 2D perovskite materials [1]. Zhou et al. believe that this phenomenon is due to the incorporation of large-sized organic cations into 2D perovskite, resulting in dynamic stability of perovskite structure and separation of external environment [13].

In summary, the dimension of 3D perovskite materials is reduced to 2D perovskite materials, and the crystal structure of halide perovskite is changed, so that the 2D perovskite formed after its octahedron is distorted has excellent stability. In other words, under certain environmental variables, 2D perovskite materials are more difficult to decompose than 3D perovskite materials.

3. The advantages of 2D organic-inorganic perovskites

The technology of perovskite has advanced rapidly over the past decade, pertaining to the domains of solar cells and LEDs, 2D perovskite materials are widely used . Compared to the 3D material perovskite. In optoelectronics, 2D material perovskite offers greater than average moisture and temperature stabilities. In contrast to conventional 3D perovskites, 2D perovskites are characterized by the insertion of organic layers between inorganic crystals [2]. Due to its alternating layered characteristics of organic-inorganic layers, the 2D perovskite material has good stability and increased exciton characteristics [2]. Additionally, engineering vertical crystal development is a key strategy for achieving effective charge transmission when the number of organic divisions rises.

In contrast to 3D perovskites, the organic cations in 2D perovskites are no longer confined to the octahedral structure. Since the first synthesis of 2D perovskite materials, various organic cations have been used to prepare 2D perovskites, including linear alkylamines and some large groups of ammonia (e.g., aniline and its derivatives, polymeric ammonia). Since the organic and inorganic 2D perovskite has weak van der Waals in the organic layer and strong ion bonds, covalent bonds in the inorganic layer, they are connected by these bonds. Related to 3D perovskite materials structures, Organic cations in 2D perovskite materials extend into a larger area and engage in van der Waals interactions. 2D perovskite has greater stability as a result.

4. The applications of organic-inorganic 2D perovskites and 3D perovskites

As early as 2007, perovskite nanosheets have been used in IC, solar cell, LEDS and other fields. Dehaven and his team applied for a patent in 2007 [14]. In this patent, they used melt-processed semiconducting organic-inorganic hybrid material to form channel layer in the field effect transistor. This technology is used for nonlinear optical/photoconductive device, flat panel display, chemical sensor, emitting and charge transporting layers in organic-inorganic LEDS, and various flexible and low-cost electronic devices. Field effect tubes made of this perovskite have many advantages. Because it is fabricated without the use of a solvent, this production method can not only make the field effect tube have better voltammetry characteristics, but also make the production cost of the field effect tube greatly reduced, and is very environmental protection.

In recent years, perovskite research results in solar cells is one of the most advanced areas. Organic-inorganic hybrid solar cells have received a lot of interest recently as a solar cell with great development prospects. However, it also has some very obvious disadvantages, such as their stability against high temperatures, light, and humidity. Device stability can be improved through ETL or HTL performance improvements, interface engineering, and active layer modifications. Stable solar cells can be made using a straightforward spin-coating procedure and a photoactive layer made of 2D perovskites. The perovskite crystals which are shielded from moisture in the atmosphere by the hydrophobic organic layer, was reported to increases the working stability of devices. Additionally, under the identical preparation conditions, 2D perovskite material films outperformed 3D analog films in terms of film formation, resulting in reduced leakage currents in electronic devices. Intercalation's organic cations prevent charge transfer., but it may be helpful to carefully control the vertical positioning of the inorganic crystals [2].

Compared with traditional solar cells, due of their benefits, perovskite solar cells have caught the interest of researchers, such as suitable band gap, large extinction coefficient, high carrier mobility of electrons and holes , and small exciton binding energy [15]. Perovskite materials can achieve photoelectric conversion efficiency of more than 23% in small area cells. In Figure 3, the structure of

a perovskite solar cell are all depicted. Since the active layer is where perovskite is most commonly employed, altering the material will have an immediate impact on the active layer's characteristics.

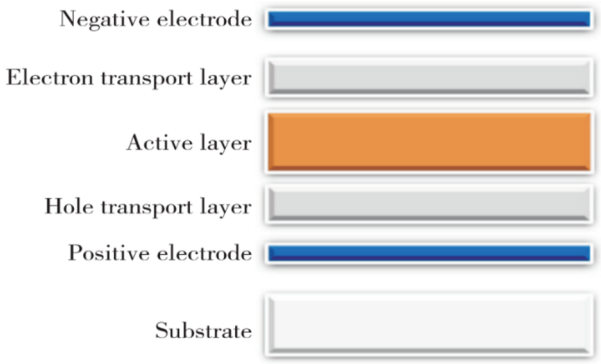


Figure 3. The PSCs' schematic diagram [17].

An Shichong et al. summarized the research progress of transparent electrodes applied to translucent perovskites and laminated solar cells [16]. At present, the highest efficiency of laminated solar cells has reached 25.5%, which has high application potential. Meanwhile, Dong Lika et al. presented the research condition of flexible and textile-based PSCs and offered suggestions for enhancing PSCs' efficiency [17].

Although 3D halide perovskite materials perform well in the experimental environment, they cannot be widely used for commercial use because of the unstable properties of ionic compounds and the presence of lead element, which are prone to decomposition after long-term use. In order to maintain the excellent photoelectric properties of 3D perovskite materials as much as possible and reduce the harm to the environment caused by its decomposition, Xiaogong Bai et al. proposed to change lead element into tin element mixed with lead element [18]. In addition, Lei Wang et al. found that FA^+ and Br^- can reduce carrier recombination [19]. For perovskite materials without limiting special components, It is thought that altering ions or ion group fractions will increase perovskite's stability and transport capabilities. Meanwhile, in an effort to advance the stability of PSCs and provide the possibility for future commercialization, heterogeneous structure has also develop into a new research fad. To increase the 3D halide perovskite materials' stability, whether by doping other elements, changing ion components or constructing heterojunctions, is currently limited by technical requirements and manufacturing costs, so it cannot be used in large areas. Therefore, the primary focus of future research will be on ways to improve stability of 3D halide perovskite materials while keeping their chemical and physical properties at cheap cost and with minimal technical requirements.

Due to its adjustable bandgap, high color purity, low non-radiative composite rate, and favorable charge transport properties, organic inorganic perovskites are also promising in electroluminescence in addition to their use in 2D perovskite materials for LEDs. Now, the brightness of perovskite LEDs has been over 10,000 cd m^{-2}. Fei Yan et al. prepared $CH_3NH_3PbBr_3$ amorphous nanoparticles as emission materials [20], with an optimal CE of 11.49 cd A^{-1}, an EQE of 3.8%. In contrast, the emission efficiency of quasi-2D perovskites is significantly higher than that of 3D perovskites, due to the self-organizing quantum wells' exciton limitation effect. [12]. With improved exciton restriction in inorganic flakes, by introducing a 2D configuration, the binding energy of perovskites can be effectively increased from tens of eVs to hundreds of eV. Strongly bound excitons in 2D perovskites typically undergo faster radiation recombination than in 3D. When the smaller diffusion length is combined, excitons are less likely to be sated by imperfections at grain boundaries and thin film surfaces, which eventually results in a comparatively high PLQE in quasi-2D perovskites. So far, quasi-2D perovskites have been used to accomplish the outstanding PLQY and EQE of LEDs devices [2].

Tan et al. reported the near-infrared and green LED device based on $MAPbBr_3$ and $MAPbI_{3-x}Cl_x$, two 3D perovskite materials [21]. Therefore, perovskite LED has attracted the attention of researchers.

External Quantum Efficiency (EQE) is one of the main performance indicators of photodetectors. Due to low EQE, 3D halide perovskite leds were not commercialized. The angular sharing $[PbX_6]^{4-}$ octahedral structure of 2D halide perovskite can accommodate large A-site cations. Meanwhile, the energy difference of van der Waals forces between inorganic layer ionic bonds and organic molecules results in the original 3D halide perovskite materials becoming layered perovskite. The alternately stacked organic-inorganic components, combined together by weak van der Waals forces, form an internal multi-quantum well structure [22]. Under the influence of quantum effect, semiconductor materials have many sub-levels in valence band and conduction band, and there are carrier transitions between these sub-levels. At the same time, electrons and holes are bound in a quantum well, which is easy to form excitons, and the exciton absorption peak appears when natural absorption occurs, thus achieving higher fluorescence efficiency. At the same time, due to the anisotropic growth characteristics of low-dimensional perovskite materials, which produces highly heterotropic photoelectric properties and large exciton binding energy, 2D perovskite materials have a wider prospect in light emitting devices than 3D perovskite materials.

Yuan prepared $(MA)_{n-1}PEA_2Pb_nI_{3n+1}$ 2D perovskite materials by using larger PEA as a cationic part instead of MA^+ [23]. This material exhibits 8.8% EQE at low voltage, which reflects the effect of high carrier migration under the characteristics of multi-quantum well structure inside 2D perovskite materials. Wentao Bi used an answer to treat the surface of 2D perovskite, thereby reducing non-radiative recombination caused by defects and improving LED efficiency [24].

Figure 4 shows the $(C_6H_9C_2H_4NH_3)_2PbI_4$ photodetector made by Ahmad et al. and its section SEM [25]. This device achieves 10% EQE under 508nm illumination. Song et al. used the 2D $CsPbBr_3$ nanosheet flexible photodetector to compare with the current Si photodetector [26], and found that due to the influence of bias, the switching ratio was greater than 104, and the response rate at 517nm could reach 0.25A/W, superior to conventional silicon-based photodetectors that are currently commercially available.

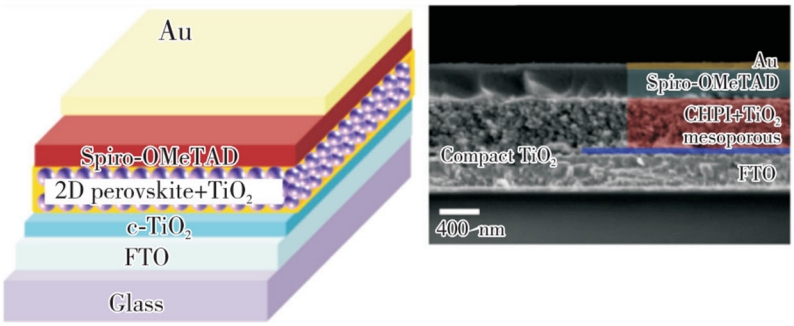


Figure 4. The schematic diagram and sectional SEM image of photodetector [25].

However, because 2D $CsPbBr_3$ nanosheets are not easy to mass produce, it is difficult to achieve large-scale application like traditional silicon-based photodetectors. Wu Jionghua et al. used the two-step mutual diffusion method to prepare high-performance $CsPbCl_3$ film UV photoelectric detector [27], and the 2D perovskite materials grown by the two-step mutual diffusion method reduced the difficulty of preparation, which provided the possibility of commercialization for photodetectors based on 2D perovskite materials

5. Conclusions

For 3D perovskite materials, it has many advantages. For example, solar cells made from three-dimensional perovskite not only have higher conversion efficiency, but also lower cost and simpler process. However, 2D perovskite materials not only have most of the excellent characteristics of 3D materials, but also are better transport characteristics, stability and so on than 3D perovskite materials due to their 2D structure. Among the wide variety of 2D perovskite materials, 2D organic-inorganic

perovskites not only make it possible to combine the superior properties of organic and inorganic materials at the molecular scale, but also provide a good opportunity to further study the relationship between structures and properties. These materials have shown good potential applications in optoelectronic devices like solar cells, field effect tubes, photodetectors, LED, and lasers. This is due to the unique structural composition characteristics of 2D perovskite and a variety of unique properties such as low synthesis cost, abundant raw materials, good charge transport, easy mass production, high crystallinity, high quantum yield, and excellent light absorption. The following aspects are anticipated to receive more attention in the future: (1) gradually improve temperature stability, photostability, oxygen and water stability of 2D perovskite materials, control defects, remove lead components, and achieve harmless materials; (2) Improve the fabrication process of 2D perovskite devices and develop more convenient technologies, such as screen printing and low temperature treatment methods; (3) The important physical processes of optoelectronic devices based on 2D perovskite materials are thoroughly discussed, such as the charge transmission direction of photodetectors, the principle of interfacial diffusion mechanism and the mechanism of device operation. (4) Develop new 2D perovskite materials with higher mobility and higher carrier density. With further investigation on perovskite, this excellent material will be used in more fields.

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