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Two-dimensional hybrid organic-inorganic perovskites as emergent ferroelectric materials

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ABSTRACT
Hybrid organic-inorganic perovskite (HOIP) materials have attracted significant attention in photovoltaics, light emission, photodetection, etc. Based on the prototype metal halide perovskite crystal, there is a huge space for tuning the composition and crystal structure of this material, which would provide great potential to render multiple physical properties beyond the ongoing emphasis on the optoelectronic property. Recently, the two-dimensional (2D) HOIPs have emerged as a potential candidate for a new class of ferroelectrics with high Curie temperature and spontaneous polarization. Room-temperature solution-processability further makes HOIP a promising alternative to traditional oxide ferroelectrics such as BaTiO₃ and PbTiO₃. In this perspective, we focus on the molecular aspects of 2D HOIPs, their correlation with macroscopic properties, as well as the material design rules assisted by advanced simulation tools (e.g., machine learning and atomistic modeling techniques). The perspective provides a comprehensive discussion on the structural origin of ferroelectricity, current progress in the design of new materials, and potential opportunities and challenges with emerging materials. We expect that this perspective will provide inspiration for innovation in 2D HOIP ferroelectrics.

I. INTRODUCTION
Ferroelectrics are distinguished by their polar crystallographic structure. Below a critical temperature (Curie temperature, \( T_c \)), these materials exhibit spontaneous polarization (\( P_s \)) which can be re-oriented under an external electric field. In general, the spontaneous polarization originates from either the ordering of pre-existent local dipoles (i.e., "order-disorder" type transition, as in the case of NaNO₂ or KH₂PO₄) or the emergence of local dipole moments induced by atomic displacement (i.e., "displacive" type transition, as in the case of traditional oxide perovskites such as BaTiO₃ or PbTiO₃). Typical ferroelectrics with switchable polarization show interesting properties such as the long-range ordered domain structure and unique hysteretic behavior, allowing numerous applications including nonvolatile memory devices, sensors, nonlinear optical devices, energy harvesters, energy storage devices, etc. Since the early report on ferroelectric Rochelle salt \([\rm KNaC_4H_4O_6(4H_2O)]\) in 1921, numerous efforts have been made to design a wide range of ferroelectric materials. To date, most research has focused on inorganic oxides with an ABO₃ perovskite structure such as PbTiO₃ and BaTiO₃ owing to their good ferroelectric performance. The processing of traditional ABO₃ ferroelectrics requires high temperature, and many of these materials contain expensive and rare-earth metal elements which could lead to additional environmental and cost issues. Hence, material
exploration for a novel class of ferroelectrics with excellent properties, such as that in ABO₃, but relatively simpler processing is of great importance for new generation of devices.

Hybrid organic–inorganic perovskites (HOIPs) are emerging as a promising candidate because of their easy processing (low-temperature and solution-synthesis capability) and huge structural and compositional design capacity. Compared to the prototype HOIPs [ABX₃, a three-dimensional (3D) crystal], two-dimensional (2D) HOIPs have a wider range of choice for the larger A-site cations, enhanced stability (due to the hydrophobicity of large A-site molecules in 2D HOIPs), and a wider range of tunability on B- and X-sites to accommodate the large A-site molecules (e.g., lead-free and all-organic HOIPs). By properly designing the composition and crystal structure, multi-property materials with inter-related optoelectronic, ferroelectric, and thermoelectric properties can be realized. In photovoltaics, HOIP-based solar cells have revealed an efficiency of over 24%. Recently, there have been various reports on the ferroelectricity of HOIPs. Nevertheless, the design rules related to a higher ferroelectric performance remains less understood. In this perspective, we discuss the prospects and challenges with a new class of 2D HOIP ferroelectrics by reviewing the prior and ongoing research, analyzing the structural origins for ferroelectricity, and discussing the potential molecular design rules through multiple simulation tools.

II. 2D STRUCTURE

Similar to their inorganic counterpart, the prototype HOIP has a three-dimensional (3D) “perovskite” structure with a general formula of ABX₃ [Fig. 1(a)], where A represents the organic molecular cation such as methylamine (CH₃NH₃), B is a metal cation such as Pb²⁺ and Sn⁴⁺, and X is halogen anions such as Cl⁻, Br⁻, and I⁻. The A-site cations are caged in a (BX₆) corner-sharing octahedral network via the hydrogen bond of “N–H…X” between the ammonium group from the A-site cation and the halogen from octahedra. For a prototype 3D HOIP, the size of the A-site cation is limited to around 2.6 Å according to Goldschmidt geochemistry rules. While for 2D HOIPs, the A-site cation can be larger and will be more choices to construct a new HOIP [Fig. 1(b)]. Specifically, when a larger organic cation (e.g., amine with long organic chains as in the case of BA⁺, butylammonium; PMA⁺, phenylethylammonium, or PEA⁺, phenylmethylammonium) is introduced into the A-site, the neighboring BX₆ octahedral layer will be separated, forming a 2D or a quasi-2D structure. In these HOIPs, each BX₆ inorganic layer is separated by those large organic A-site spacers. The organic and inorganic layers are held together by hydrogen and Coulombic interactions, whereas neighboring organic layers are held via van der Waals interaction [Fig. 1(c)]. Besides, the nucleophilic BX₆ inorganic layer, associated with the halogen atom (X), might also interact with the electrophilic organic layers via halogen bonding (electrostatic interaction due to the anisotropy of the electron density around the organic halogen nucleus). The hydrogen bonding and the halogen bonding that “anchor” the organic spacer to the inorganic layers are relatively weak, which tend to break at elevated temperatures and enable the reorientation of organic spacers. A formula of R₂A⁻₁BₓX₃n⁺₁ is typically used to describe the 2D and quasi-2D perovskite, where R is the large organic cation (e.g., an aromatic or aliphatic alkylammonium) introduced as a spacer that separates the inorganic layers and index n refers to the number of inorganic octahedral unit sheets that are held together. When n = 1, a single sheet of inorganic BX₆ octahedra is separated by an organic layer comprising of the organic spacer (R cation in R₂A⁻₁BₓX₃n⁺₁), which is a “strict 2D” HOIP. If n > 1, a relatively smaller A-cation fills the voids in the inorganic layer and induces the formation of “quasi-2D” HOIP. In this case, there are n sheets of an inorganic octahedral unit separated by the organic spacers. Eventually as n increase to infinity, the structure converges to a typical 3D HOIP configuration.

The unique large asymmetric A-site cation offers an additional asymmetry to the 2D HOIP crystal, the alignment of which gives rise to the ferroelectricity (in accordance with the classic “order-disorder” type ferroelectric). In comparison, 3D HOIPs have a relatively lower ferroelectric response as the property mainly arises from the displacement of the smaller A-site cation, following the classic “displaceable” type transition. Such a large A-site space in 2D HOIP thus gives a large molecular design capacity, where complex A-site molecules with large dipole moments can be used to enhance the polarization. The weak hydrogen and halogen interactions between these A-site organic spacers and BX₆ octahedra layer allow a certain level of A-site dynamics, allowing the A-site dipole ordering and a resultant polarization. 2D HOIPs usually have a multiple-quantum-well structure, where the A-site organic spacer has a layer thickness of more than 10 Å [e.g., 13.88 Å in (C₄H₉NH₃)PbI₄ 2D HOIP] and the BX₆ octahedral layer is of several angstroms [e.g., 6.32 Å in (C₄H₉NH₃)PbI₄ 2D HOIP]. In such a unique spatial confining system, the organic spacers with restricted movement tend to self-align their dipole moments in an ordered configuration at low temperatures. At higher temperatures, organic spacers are disordered, thereby, diminishing the original polarization. Such behavior is analogous to the ferroelectric transition in many “order-disorder” type oxides such as NaNO₂.

Through the design of the A-site spacer in terms of molecular dipole moment, molecular size, interaction with the BX₆ octahedral layer, the 2D HOIPs offer a good platform to investigate, design, and develop a new “order-disorder” type halide ferroelectrics. Prior success has revealed some exciting results on 2D HOIPs with significantly larger polarization [P = 13 μC cm⁻² in (benzylationmmonium)₂PbCl₄] than 3D HOIPs [from 2 μC cm⁻² in Fe-doped MAPb₀.₇Fe₀.₃I₃ reaching up to 6.8 μC cm⁻² in (MeHdabc)RbI₃]. In addition to ferroelectric performance, another advantage of 2D HOIPs is their stability as the alkyl groups present in the large A-site spacers in 2D HOIPs are more hydrophobic than those in 3D HOIPs. The ionic motion in typical 3D HOIPs is significantly restricted in comparison to 2D HOIPs. These results indicate that 2D HOIPs have promising potential for both fundamental studies and practical applications.

III. STATE-OF-THE-ART MATERIALS

A. 2D HOIP ferroelectrics

2D HOIP ferroelectrics have a chemical formula of RₓBX₄, where the interlayer of large organic spacers (R) is inserted between the BX₄ corner-sharing octahedral layer. The ordering of dipole
FIG. 1. Structure of 3D and 2D HOIP materials. (a) Illustration of 3D HOIP ABX₃ showing an A-site cation in voids of a corner-sharing BX₆ octahedral. (b) Choice of A, B, and X site elements in the 3D structured HOIP. (c) Illustration of low dimensional HOIP structure with different layers of perovskite (n). The 2D perovskite has one single octahedral layer (n = 1) with a formula of R₂BX₄. R is a large organic cation. Some example R cation are listed: BA⁺ = butylammonium, PMA⁺ = phenylmethylammonium, PEA⁺ = phenylethylammonium. For n > 1, the quasi-2D HOIP contains multiple octahedral layer and the corresponding formula is R₂An−₁BnX₃n+1. Data in Fig. 1(b) are adapted with permission from Wang et al., Prog. Mater. Sci. 106, 100580 (2019). Copyright 2019 Elsevier B.V. Figures 1(a) and 1(c) were adapted with permission from Grancini and Nazeeruddin, Nat. Rev. Mater. 4, 4–22 (2019). Copyright 2019 Springer Nature.
moment from the organic spacers results in the polarization, and
the disordering at higher temperatures results in the transition
between ferroelectric and paraelectric. Building upon this mecha-
nism, various ferroelectric 2D HOIPs have been developed. The 2D
HOIP (Bza)2PbCl4 (Bza = benzylammonium) has a room tempera-
ture phase belonging to the polar Cmc21 space group (point group
mmm2) as recorded in the Cambridge structural database (reference
code: HORFAV), which indicates that it is a potential ferroelectric
material.34 Liao et al.35,36 reported ferroelectricity in (Bza)2PbCl4 in
2015. From the polarization point of view, room temperature struc-
ture is characterized by the ordered alignment of the Bza+ cation
with all the C–N bonds in the molecules aligned along with the
crystallographic c axis as illustrated in Fig. 2(a). Thus, the sponta-
neous polarization mainly lies along the c axis. With temperature
surpassing the transition temperature Tc, the A-site shows rota-
tional disordering with the rotation center axis perpendicular to
the a−c plane. As a result, the crystal structure evolves from Cmc21
to a nonpolar Cmca space group and the material becomes
paraelectric. Experimentally, the temperature-dependent dielectric
constant of the (Bza)2PbCl4 showed a characteristic change along
the c axis with a peak of 600, verifying the ferroelectricity along the
c axis with a Tc of 438 K [Fig. 2(b)]. Additionally, the characteristic

FIG. 2. (a) Illustration of the structural evolution of (Bza)2PbCl4 during ferroelectric phase transition. Figure is adapted with permission from Xu et al., Coord. Chem. Rev. 387, 398–414 (2019). Copyright 2019 Elsevier B. V. (b) Temperature-dependent dielectric measurement of (Bza)2PbCl4 along the c axis. The inset shows the P–E hysteresis loop of (Bza) measured along the c axis direction. (c) Temperature-dependent measurement of SHG coefficient χ(2) and spontaneous polarization. The inset shows the photoimage of a (Bza)2PbCl4 single crystal. Figures are adapted with permission from Liao et al., Nat. Commun. 6, 1–7 (2015). Copyright 2015 Nature.

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polarization–electric field ($P-E$) and temperature-dependent second harmonic generation (SHG) measurement further confirm the ferroelectricity along the $c$ axis [Fig. 2(c)]. It should be noted that (Bza)$_2$PbCl$_4$ shows a notable $P_S$ value of 13 μC cm$^{-2}$ at room temperature and a high $T_C$ of 438 K, comparable to widely used oxide ferroelectrics such as BaTiO$_3$ ($T_C = 390$ K, $P_S = 26$ μC cm$^{-2}$ obtained in a single crystal sample). Similarly, by tuning the A-site molecules, more 2D HOIP ferroelectrics have been investigated. Ye et al. reported the (CHA)$_2$PbBr$_4$ (CHA = cyclohexylammonium) 2D HOIP ferroelectric exhibiting a similar structural and corresponding ferroelectric transition. The material exhibits a $T_C$ around 363 K and a $P_S$ value of 5.8 μC cm$^{-2}$ at room temperature. In the low-temperature phase, all the CHA$^+$ cations have the same orientation belonging to a polar group of Cmc$_21$. At elevated temperatures above $T_C$, the CHA$^+$ experiences rotational disorder and occupies two possible orientational states related by the twofold axis along the crystallographic $a$ axis direction. Figure 3(a) shows the model of the high-temperature phase, representing the average configuration of two CHA$^+$ cation orientations. As a result, at a higher temperature, the disorder HOIP exhibited a centrosymmetric phase (Cmca). Using such a structural model, the calculated spontaneous polarization is 5.1 μC cm$^{-2}$ which matches well with the experimental result of 5.8 μC cm$^{-2}$. As the major ferroelectric contribution in these 2D HOIPs comes from the molecular dipole of the $A$-site spacer, modifying the halide composition of the inorganic layer [e.g., iodide doping in (CHA)$_2$PbBr$_{3.55}$I$_{0.45}$ and (CHA)$_2$PbBr$_{3.3}$I$_{0.7}$] did not change the ferroelectric nature. Chen et al. developed another ferroelectric 2D HOIP, (ATHP)$_2$PbBr$_4$ (ATHP = 4-aminotetrahydropyran), with a ferroelectric phase of Cmc$_21$. The orientational ordering of ATHP$^+$ organic spacers at room temperature contributes to a $P_S$ of 5.6 μC cm$^{-2}$ along the $c$ axis, which is comparable to that of (CHA)$_2$PbBr$_4$. Interestingly, this (ATHP)$_2$PbBr$_4$ exhibits a significantly larger $T_C$ of 504 K than that of (CHA)$_2$PbBr$_4$ (363 K), due to the more compact crystal packing of (ATHP)$_2$PbBr$_4$ that leads to restricted movement of ATHP$^+$ cations with temperature. Overall, the dynamic and molecular dipole from the large organic spacer is of great importance in realizing the ferroelectricity in 2D HOIPs.
Further enlarging the complexity of the A-site spacer will induce the structural evolution from 2D toward 1D. Several studies have been conducted on 1D HOIPs using larger A-site molecules and theoretical estimates have revealed an even higher theoretical $P_s$ in 1D HOIPs [e.g., $P_s = 25.6 \mu C \text{cm}^{-2}$ for diethylmethyl(2-fluoroethyl) ammonium-MnCl$_4$]. As there is less confinement effect in 1D HOIPs, there is a higher degree of freedom for the dynamic motion of A-site cations in the low temperature phase, making them typically disordered and paraelectric.

Since the organic cation spacers serve the core role in the ferroelectricity of 2D HOIP, designing the A-site organics with enlarged molecular dipole can provide higher ferroelectricity. One strategy is the H/F substitution, i.e., replacing hydrogen by fluorine to enlarge the dipole moment. Such a molecular design strategy is especially attractive in ferroelectric 2D HOIP since the hydrogen and fluorine have a similar steric parameter which ensures that such substitution on the organic cation spacer will not introduce large structural distortion and thus it can minimize the risks of unexpected structural disorder. Furthermore, the molecular dipole moment of the organic cation can be greatly enhanced owing to the strong electronegativity of fluorine, which can enhance the lattice polarization and induce a large $P_s$. The fluorination strategy of organic spacers has been used to explore new 2D HOIPs with outstanding optoelectronic properties, including (fluoroethylammonium)$_2$PbCl$_4$ and the fluorinated derivatives (phenethylammonium)$_2$PbI$_4$. Recently, Shi et al. employed this molecular design strategy to fabricate new ferroelectric 2D HOIP with high performance based on fluorinated aromatic cations fluorobenzylammonium (FBA). The material (2-FBA)$_2$PbCl$_4$ shows a similar ferroelectric phase transition as that of its non-fluorinated counterpart (benzylammonium)$_2$PbCl$_4$, with the symmetry change from a polar Cmc2$_1$ to a nonpolar I$_4/mmmm$ group across a $T_C$ of 448 K, due to the orientational disordering of FBA$^+$ [Fig. 3(b)]. The dipole moment of the C–F bond ($\sim$2 D) is much higher than the original C–H bond (1.4 D). Through alignment, the integrated dipole moment will lead to a considerable contribution to the total polarization. As a result, the polycrystalline (2-FBA)$_2$PbCl$_4$ sample shows a high $T_C$ of 448 K and $P_s$ of about 5.35 $\mu C \text{cm}^{-2}$, which is comparable to $P_s$ of the single-crystalline (BA)$_2$PbCl$_4$ sample. These results demonstrate that the fluorination could be an efficient molecular design strategy for organic cation spacers to develop high-performance 2D HOIP ferroelectrics. However, the fluorine substitution site should be carefully considered, as the authors also found that the material shows different symmetry when fluorine substitution occurs at different positions on the benzene ring in the A-site spacer. The (4-FBA)$_2$PbCl$_4$ and (3-FBA)$_2$PbCl$_4$ crystallize into centrosymmetric Pmma and P2$_1$/c space groups, respectively, and these materials lose their ferroelectricity. The molecular design strategy of H/F substitution has also been employed to create some new 2D HOIP ferroelectrics. Zhang et al. replaced the piperidinium cation (PD)$_2^+$ in a non-ferroelectric HOIP (PD)$_2$PbI$_4$ with a difluorinated cation 4,4-difluoropiperidinium (4,4-DFPD). The introduction of fluorine changes the molecular dipole of organic cations triggering the occurrence of ferroelectricity in the material. As a result, the obtained (4,4-DFPD)$_2$PbI$_4$ shows ferroelectricity with a $T_C$ of 429 K and a large $P_s$ of 10 $\mu C \text{cm}^{-2}$. The low-temperature ferroelectric phase accommodates an Aba2 polar space group, which transitions into the paraelectric phase with a centrosymmetric I4/mmm space group above $T_C$.

Incorporation of chirality on the organic cation is another efficient way for designing 2D HOIP ferroelectrics. Homochiral molecules will form enantiomorphic crystals with the same handedness. Compared with the achiral molecules, the homochiral molecules tend to crystallize into one of the five polar enantiomorphic point groups, which enables the ferroelectricity. Such a protocol has been employed in constructing molecular ferroelectrics such as (R)-hydroxyquinuclidinium halides. Building upon this principle, Yang et al. employed homochiral cations of [R- and S-1-(4-chlorophenyl)ethylammonium]$^+$(R-CEA) and (S-CEA)$_2$PbI$_4$. Both the compounds crystallize into the P1 polar space group in the ferroelectric phase at 293 K and transition into a nonpolar P422 space group in the paraelectric phase above $T_C$. These two enantiomorphic structures show slightly different $T_C$ of 483 K and 473.2 K for (R-CEA)$_2$PbI$_4$ and (S-CEA)$_2$PbI$_4$, respectively. Interestingly, the racemic counterpart of these two enantiomorphic structures adopts a nonpolar P2$_1$/c space group at room temperature and shows no ferroelectric phase transition. Therefore, incorporation of homochiral organic molecules might facilitate crystallization of 2D HOIP into polar space groups. This finding suggests an efficient way for designing new 2D HOIP ferroelectrics by incorporating chirality in the organic cations.

In summary, the order–disorder orientational movement of organic cations plays the dominant role in ferroelectric–paraelectric transition in most of the 2D HOIPs discovered so far. Therefore, new 2D HOIP ferroelectrics can be explored by the systematic molecular design of the organic cations through the modification of the composition or geometry via strategies such as H/F substitution and incorporation of chirality.

B. Quasi-2D HOIP ferroelectrics

The quasi-2D HOIP has a more complex structure compared to the 2D HOIP and typically has a formula of R$_2$An–I$_{BnX_{3n}+1}$. In addition to the large organic cation spacer separating the inorganic layers, there are additional small A-site cations enclosed in the voids of the octahedral sheets. The structural temperature-dependent phase transition in these quasi-2D HOIPs is more complex compared to the 2D HOIPs, as the smaller A-site cations enclosed in voids of the octahedral sheets and distortion of octahedra will provide an additional contribution to polarization through lattice displacement. Li et al. reported the ferroelectric multilayered (n = 3) quasi-2D HOIP (BA)$_2$(MA)$_n$Pb$_{3n}$Br$_{6n}$ (BA = n-butylammonium, MA = methylammonium, CH$_3$NH$_3$) by alloying the 3D HOIP MAPb$_{3n}$Br$_{6n}$ perovskite with n-butylammonium. The electric polarization is induced by the synergic order-disorder transition of organic components. At room temperature, this material crystallizes into a ferroelectric phase with a polar space group of Cmc2$_1$, as revealed by the single-crystal x-ray diffraction analysis. The MA$^+$ cations are enclosed in the cavities of corner-sharing PbX$_6$ octahedra, while the BA$^+$ cations serve as organic spacers to separate the tri-layered...
inorganic framework, as shown in Fig. 4(a). The MA$^+$ cations are ordered and oriented along the $c$ axis, while the BA$^+$ organic spacers also have ordered arrangement, which induces the polarization. Besides, the inorganic framework shows structural distortion with a small-angle tilt deviating from its octahedral symmetry. As a result, the Pb$^{2+}$ ion moves away from the center of the PbBr$_6$ octahedra, which also contributes to the electrical polarization. At elevated temperatures, above $T_C$ ($T_C = 315$ K), both the MA$^+$ and BA$^+$ become highly disordered and the configuration of the inorganic framework becomes more symmetric, hence the material transitions into a paraelectric phase with a centrosymmetric space group of $Cmca$. The material shows a $P_S$ value of $3.5 \mu C cm^{-2}$ at $307$ K. Based on the same material system and similar concept, Li et al.55 used FA (FA = formamidinium, NH$_2$CHNH$_2$) as the A-site cation in an octahedra framework and prepared a new bi-layered ($n = 2$) quasi-2D perovskite of (BA)$_2$(FA)Pb$_2$Br$_7$. Below $T_C$, the material has a polar ferroelectric phase of $Cmc2_1$. The distortion of PbBr$_6$ octahedra and ordering of BA$^+$ organic spacers induces the displacement of negative and positive charge centers along the crystallographic $c$ axis, thus generating dipole moments and electric polarization along this direction. At high temperatures, the material shows a paraelectric phase with a nonpolar space group of $Cmcn$ as a result of BA$^+$ disordering and symmetrical arrangement of PbBr$_6$ octahedra. The material shows a moderate Curie temperature of $322$ K and a sufficiently high $P_S$ value of $3.8 \mu C cm^{-2}$. In a similar system of (BA)$_2$(MA)$_{n-1}$PbnBr$_{3n+1}$, Wu et al.56 replaced the MA$^+$ with inorganic element Cs$^+$ when $n = 2$ to fabricate (BA)$_2$CsPbBr$_7$ as a novel quasi-2D HOIP ferroelectric. This material undergoes a ferroelectric transition at $412$ K, with the symmetry change from a polar space group of $Cmc2_1$ to a nonpolar $Cmca$. The structural transition is slightly different from (BA)$_2$(MA)$_3$Pb$_3$Br$_{10}$. At room temperature, in addition to the ordering of the BA$^+$ organic spacers along the $c$ axis, the introduced Cs$^+$ atom shows distinct displacement away from the center of octahedra cavities along the $c$ axis. As a result of these motions, the positive and negative charge centers separate, which induces dipole moments and spontaneous polarization along the crystallographic $c$ axis direction as illustrated in Fig. 4(b). At a temperature above $T_C$ ($T_C = 412$ K), the BA$^+$ organic spacers transform into a disordered state with two possible orientations associated with a mirror plane, while the Cs$^+$ atoms reside at the center of octahedra cavities. Thus, the material becomes centrosymmetric exhibiting negligible polarization. The measured $P_S$ has a value of $4.2 \mu C cm^{-2}$ at $344$ K, demonstrating the effective contribution of polarization from the displacement of Cs$^+$. In a summary, compared with the 2D HOIPs, the quasi-2D HOIPs have a more complicated ferroelectric phase transition mechanism due to the involvement of small A-site cations. Besides the organic spacers, the displacement of small A-site cations and associated distortion of the B$_n$X$_{3n+1}$ octahedral layer will give an additional contribution to the ferroelectric polarization. Hence, the quasi-2D HOIP ferroelectrics are mixed type consisting of...
“order-disorder” and “displacive” nature. Quantifying contribution from each mechanism to the overall ferroelectricity is important in optimizing the molecular design for new quasi-2D HOIP ferroelectrics.

IV. APPLICATIONS AND CHALLENGES

So far, various 2D HOIP ferroelectrics have been developed, displaying a wide range of performance. Table I summarizes the ferroelectric properties and structural phase transition of reported 2D HOIP ferroelectrics and their comparison with traditional oxide ferroelectrics. The 2D HOIPs show sufficiently large spontaneous polarization. Particularly, all these materials show a high \( T_C \) above the room temperature, and some pure 2D HOIPs displays a \( T_C \) larger than the commonly used BaTiO\(_3\),\(^{35}\) approaching that of PbTiO\(_3\).\(^{58}\) The high \( T_C \) and solution-based processability are promising for the broad application of 2D HOIP in various room-temperature ferroelectric devices.

In terms of applications, one early motif for developing new ferroelectric HOIPs is to further improve the efficiency of photovoltaic devices through the photo-ferroelectric effects.\(^{59}\) It has been proposed that the ferroelectric domains in HOIPs provide channels for photocarrier transport as the electrons and holes can move along the potential maxima and minima at the antiphase boundary,\(^{60}\) which can reduce the unwanted carrier recombination loss.\(^{61}\) Furthermore, the presence of ferroelectric domains is also
The HOIPs can be easily processed in the form of a solution to produce single crystals or polycrystalline films. Such an ease of production allows the fabrication of thin-film energy harvester based on the direct piezoelectric effect. So far, there is limited progress using the 2D HOIPs for energy harvesting since most of the studies focus on piezoelectric harvesters based on 3D HOIPs. For instance, Kim et al. demonstrated a flexible piezoelectric generator using a compositionally engineered MAPbI3 film, where Pb2+ was partially substituted by Fe2+. The device based on MAPb1−xFexI3 (Figs. 5(d)–4(f)), which exhibited high performances including an extremely low dark current of several pA, a large on/off current ratio ~2.5 × 105, and an ultrafast response rate of 150 ms. This superior photodetection performance benefits from the ultrahigh electrostatic field enabled by ferroelectric polarization, which greatly suppresses the dark current. Moreover, the 2D HOIP also shows excellent nonlinear optical properties compared with their inorganic counterpart. Recently, Li et al. discovered a giant two-photon absorption coefficient of 5.76 × 103 cm GW−1 in ferroelectric (BA)2(FA)PbBr4. This value is nearly two-orders of magnitude higher than that of the conventional inorganic perovskite ferroelectrics such as BaTiO3 and Ce:BaTiO3. Such prominent two-photon absorption performance is not only desirable for nonlinear photonic application but also promising for vis–IR dual-modal light harvesting or photo-sensor device.

Another potential application of HOIPs would be the ferroelectric memory device. Several signs of progress have been made toward resistive switching memories. The resistive switching behavior in HOIP is thought to link to the ferroelectric polarization switching. The material can switch among two different electrical

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### TABLE I. Phase transitions and P \(_{\text{r}}\) of 2D HOIPs compared with conventional inorganic perovskite.

<table>
<thead>
<tr>
<th>Compound</th>
<th>(T_c) (K)</th>
<th>(P_{\text{r}}) (μC cm(^{-2}))</th>
<th>Symmetry transition</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure 2D HOIP</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Bza)(_2)PbCl(_4)</td>
<td>438</td>
<td>13</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>35</td>
</tr>
<tr>
<td>(2-FBA)(_2)PbCl(_4)</td>
<td>448</td>
<td>5.4 (polycrystalline film)</td>
<td>Cmc2(_1) ↔ 4/mmm</td>
<td>50</td>
</tr>
<tr>
<td>(CHA)(_2)PbBr(_4)</td>
<td>363</td>
<td>5.8</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>37</td>
</tr>
<tr>
<td>(ATHP)(_2)PbBr(_4)</td>
<td>503</td>
<td>5.6</td>
<td>Cmc2(_1) ↔ N.A.</td>
<td>38</td>
</tr>
<tr>
<td>(4,4-DFFPD)(_2)PbI(_4)</td>
<td>429</td>
<td>10</td>
<td>Aha2 ↔ 4/mmm</td>
<td>51</td>
</tr>
<tr>
<td>(R-CEA)(_2)PbI(_4)</td>
<td>483</td>
<td>14</td>
<td>P1 ↔ P422</td>
<td>53</td>
</tr>
<tr>
<td>(S-CEA)(_2)PbI(_4)</td>
<td>473</td>
<td>14</td>
<td>P1 ↔ P422</td>
<td>53</td>
</tr>
<tr>
<td>(4,4-DFFHA)(_2)PbI(_4)</td>
<td>454</td>
<td>1.1</td>
<td>P2(_1) ↔ Cmc2(_1) ↔ 2/mmm ↔ I42 m</td>
<td>27</td>
</tr>
<tr>
<td>(BA)(_2)PbCl(_4)</td>
<td>328</td>
<td>2.1</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>128</td>
</tr>
<tr>
<td>(EA)(_2)FeCl(_4)</td>
<td>98</td>
<td>N.A</td>
<td>P2(_1) ↔ Pbca</td>
<td>129</td>
</tr>
<tr>
<td>(Bza)(_2)MnBr(_4)</td>
<td>421</td>
<td>2.3</td>
<td>P2(_1) ↔ N.A.</td>
<td>130</td>
</tr>
<tr>
<td>Quasi-2D HOIP</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(BA)(_2)(MA)(_2)Pb(_2)Br(_10)</td>
<td>315</td>
<td>2.9</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>54</td>
</tr>
<tr>
<td>(BA)(_2)(FA)PbBr(_7)</td>
<td>322</td>
<td>3.8</td>
<td>Cmc2(_1) ↔ Cmcm</td>
<td>55</td>
</tr>
<tr>
<td>(BA)(_2)CsPb(_2)Br(_7)</td>
<td>412</td>
<td>4.2</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>56</td>
</tr>
<tr>
<td>(BA)(_2)(MA)Pb(_2)Br(_7)</td>
<td>352</td>
<td>3.6</td>
<td>Cmc2(_1) ↔ Cmca</td>
<td>131</td>
</tr>
<tr>
<td>(R3HQ)(_2)KCe(NO(_3))(_8)</td>
<td>315</td>
<td>4.0</td>
<td>P2(_1) ↔ N.A.</td>
<td>132</td>
</tr>
<tr>
<td>(R3HQ)(_2)RbEu(NO(_3))(_8)</td>
<td>347</td>
<td>N.A.</td>
<td>P2(_1) ↔ N.A.</td>
<td></td>
</tr>
<tr>
<td>(R3HQ)(_2)RbSm(NO(_3))(_8)</td>
<td>350</td>
<td>N.A.</td>
<td>P2(_1) ↔ N.A.</td>
<td></td>
</tr>
<tr>
<td>(R3HQ)(_2)RbTb(NO(_3))(_8)</td>
<td>340</td>
<td>N.A.</td>
<td>P2(_1) ↔ N.A.</td>
<td></td>
</tr>
<tr>
<td>Inorganic perovskite</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BaTiO(_3)</td>
<td>393</td>
<td>26</td>
<td>P4 mm ↔ Pm3 m</td>
<td>52</td>
</tr>
<tr>
<td>PbTiO(_3)</td>
<td>763</td>
<td>50</td>
<td>P4 mm ↔ Pm3 m</td>
<td>53</td>
</tr>
</tbody>
</table>
states under different bias conditions: low-resistance state (LRS) and high-resistance state (HRS), which corresponds to the “ON” and “OFF” states to be used for data memory. At present, the memory device based on lead-free HOIP, (MA)\(_3\)Bi\(_4\)I\(_9\)\textsuperscript{85} [Figs. 5(g)–4(f)] has shown promising performance with good retention property of 10\(^4\) s, switching speed of 100 ns, and endurance over 300 cycles, as well as environmental stability. These results demonstrate the potentials of HOIP as a candidate for next-generation, memory storage devices. These prior results have shown the multi-physical properties of the HOIPs which inspire researchers to use HOIPs beyond their popular photovoltaic applications. Bearing the unique structural and ferroelectric property compared with their 3D counterparts, the 2D HOIPs may have large potential for photodetectors, nonlinear optics, piezoelectric energy harvesting, and nonvolatile memory devices. Moreover, the intrinsic environmental stability of 2D HOIPs makes them more suitable for practical device applications.

As a relatively new field, there is a large space in an exploration of the ferroelectric 2D HOIPs. (1) First, although more and more 2D HOIPs with notable ferroelectric performance are discovered, there has not been a universal mechanism to describe the polarization in these materials. Prior researchers highlight the importance of polarization contribution from organic spacer alignment below \(T_c\) but have not fully revealed the correlation of the molecular dipole of organic cation spacer and their orientation alignment to the spontaneous polarization. The polarization mechanism is even more complex in the quasi-2D HOIP \((R_3A_{n-1}X_{n+1})\) as there could be contributions from the inorganic framework as well as the smaller A-site organic cation, making it more challenging in controlling the overall ferroelectric property. The current design of ferroelectric 2D HOIP is largely based on “trial and error” due to the lack of theoretical guidance, which hinders the exploration of new ferroelectric HOIPs. Therefore, a comprehensive study relating the polar property of organic cations, their evolution of orientational configuration during phase transition, and the ferroelectricity of 2D HOIP is highly desired. (2) Second, although the newly discovered 2D HOIPs show considerably high \(T_c\), their spontaneous polarization value is still inferior to the widely used inorganic perovskite of BaTiO\(_3\) and PbTiO\(_3\). Strategies are needed to enhance the polarization strength in 2D HOIP. As previously presented, H/F substitution could be an efficient way to enhance the molecular dipole of organic cation spacer and ferroelectric polarization of the 2D HOIP. Other strategies for molecular engineering including manipulating the stereographic geometry of the organic cations and incorporation of chirality might also be considered to tune the polar property of organic cation and improve the ferroelectric property of 2D HOIP. (3) Third, practical demonstration of 2D HOIPs in electronic devices is still lacking compared with their 3D counterparts, which might be due to the difficulties in achieving high-quality thin films. New methods are needed to produce large-size, high-quality, uniform 2D HOIP crystals for the future development of practical ferroelectric devices. Considering the excellent structural and compositional tunability of 2D HOIPs, there is still a large potential to design and construct new ferroelectrics with a promising magnitude of polarization and \(T_c\). Advanced computational tools such as machine learning (ML) and atomistic-scale modeling (application of artificial intelligence-enabled materials discovery and synthesis) are effective tools in accelerating the pace for design, development, and fabrication of new 2D HOIP ferroelectrics.

V. ARTIFICIAL INTELLIGENCE-ENABLED MATERIALS DISCOVERY AND SYNTHESIS

As the choice for A-site has a large number of potential candidates and the coordinated possibilities for B- and X-sites are also large, there is a huge design space for new 2D HOIP ferroelectrics. Finding the proper material composition remains a difficult task by relying on traditional “trial and error” methodologies. Building upon the fundamentals and design rules as discussed above, and utilizing the advanced computational modeling techniques, a more precise prediction of the molecular structure and material property can be realized. Emerging artificial intelligence-based algorithms that combine machine learning and atomistic-scale simulation is particularly relevant in this regard. In comparison to the traditional material development procedure that requires many years to decades to find the optimum material,\textsuperscript{85} more efficient strategy such as materials genome initiative (MGI) has produced substantial advances, in theory, modeling, simulation, computing, algorithms, software, data analysis, and experimental techniques, and digital infrastructure for sharing of data, models, and tools.\textsuperscript{72,73,74} Rapid advances in machine learning algorithms and software, together with the increasing availability of materials data, through databases such as NOMAD,\textsuperscript{75} QOMD,\textsuperscript{76} AiiDA,\textsuperscript{77} and AFLLOW,\textsuperscript{78} offers the possibility of dramatically expediting the discovery, design, and synthesis of new materials.\textsuperscript{79} For example, predictive models trained using machine learning on such databases have been used to predict materials properties, e.g., electronic bandgap from composition alone,\textsuperscript{80} and thermodynamic stability from DFT calculations\textsuperscript{81} among others.\textsuperscript{82} This opens up the possibility of using such predictive models for materials discovery to screen candidate compositions with respect to the desired properties in novel 2D HOIPs. Generative models, e.g., generative adversarial neural networks, trained on large materials datasets can be used to postulate new material compositions.\textsuperscript{83} Neural networks trained using machine learning on databases of calculated properties of materials spectra, e.g., those obtained using x-ray absorption spectroscopy, have been shown to be useful in characterizing structural changes in materials,\textsuperscript{84} classifying the specific chemical environments within samples\textsuperscript{85} in identifying sub-nanometer atomic assemblies\textsuperscript{86,87} and oxidation states etc.\textsuperscript{86} This is particularly helpful in identifying new ferroelectrics among thousands of possibilities in 2D and quasi-2D HOIP family with promising polarization and Curie temperature.

Of particular interest in the context of this perspective are the recent successes of machine learning in the discovery, design, and synthesis of perovskite materials. Pilania et al.\textsuperscript{88,89} have used a support vector machine classifier trained on a dataset of 185 experimentally known ABX\(_3\) compounds to predict viable ABX\(_3\) halide compositions (where A and B represent monovalent and divalent cations, respectively, and X is F, Cl, Br, or I anion) in the perovskite crystal structure. Pilania et al.\textsuperscript{88} and Balachandran et al.\textsuperscript{89} applied machine learning algorithms to the database of experimentally
reported $\text{ABO}_3$ compounds to predict possible new compositions. Li et al.\textsuperscript{92} used machine learning to predict the thermodynamic stability of perovskite oxides. Zhai et al.\textsuperscript{93} used machine learning to predict the Curie temperature of perovskite materials. Balachandran et al.\textsuperscript{91} trained a classifier to screen for perovskite compositions and used it to drive active learning to identify promising perovskites for synthesis and experimental evaluation. Odabasi et al.\textsuperscript{94} have applied machine learning to a database of performance data of 1921 solar cell devices extracted from 800 articles on the (organo)-lead-halide perovskite cells published between 2013 and 2018 to develop a predictive model for cell performance from fabrication conditions, device architecture, etc. Sun et al.\textsuperscript{95} have demonstrated the potential of machine learning to speed up the discovery and synthesis of novel perovskite inspired compounds by at least an order of magnitude. Howard et al.\textsuperscript{96} have argued for the use of machine learning to optimize operating parameters of perovskites to maximize their long-term power conversion efficiency.

The aforementioned advances underscore the promise and potential of machine learning to accelerate the discovery, characterization, design, modeling, and fabrication of HOIPs in general and 2D HOIPs in particular. For example, Lu et al.\textsuperscript{97} have used a combination of machine learning and DFT calculations to rapidly select, from a large number of candidates, a small number of orthorhombic lead-free HOIPs with proper bandgap for solar cells and room temperature thermal stability. The recent availability of a large HOIP dataset\textsuperscript{97} offers the possibility of applying machine learning to accelerate, perhaps by multiple orders of magnitude, the development, and fabrication of 2D HOIPs with novel ferroelectric properties.

VI. ATOMISTIC-SCALE SIMULATIONS ON HYBRID ORGANIC/INORGANIC MATERIAL INTERFACES

A combination of machine learning and artificial intelligence techniques with atomistic computational models opens promising
possibilities. Illustrating the interplay between the organic and inorganic layers at the interfaces (e.g., distortion of octahedra, reorientation of organic cation), and the resultant change in dipole moment is critical toward understanding the ferroelectric nature in both the 2D and quasi-2D HOIPs. In order to determine the structure and chemical events for complex material interfaces, like hybrid organic/inorganic materials and their interfaces, we need atomistic-scale models that describe both the physical and chemical interactions between organic and inorganic materials and can also simulate key diffusion chemical aging steps defining the formation and long-term preservation of these materials and interfaces. To provide such models, we need an atomistic-scale tool that enables us to perform relatively large (>1000 atoms), relatively long-time (> nanosecond) molecular dynamics (MD) simulations and/or mixed Monte Carlo/MD simulations. While ab initio-based methods, like DFT, provide an accurate description of reaction energies and barriers—and as such could, in principle, provide this information, the computational cost of these methods restricts their application to relatively small systems (typically <1000 atoms) and short time-scales (typically <100 ps). However, the HOIP materials, and the 2D HOIPs, in particular, usually contain organic cations with complex composition and steric structure which involves a huge number of atoms, making it even more complex to perform traditional ab initio-based simulations. To provide a comprehensive mechanism, modeling of larger systems and longer time scales is essential.

Tight-binding DFT (DFTB)90,99 and empirical reactive force field methods100,101 provide appropriate methods here, as both these methods have demonstrated the capability to accurately reproduce DFT-based reaction profiles while providing significant transferability—addressing a wide range of material types (covalent, ionic, metallic, and ceramic) and elements. Both methods are several magnitudes faster than DFT-methods (DFTB roughly 5 magnitudes, reactive force fields 6–8 magnitudes), enabling MD applications to >1 000 000 atoms and >10 ns102–104—providing sufficient size- and time ranges for studying the structure and dynamics of complex inorganic–organic interfaces. DFTB methods have the advantage of direct access to electronic properties like band spectra and electric conductivity—properties which are typically not accessible for empirical reactive force field—since these methods coarse-grain the electronic terms with functional forms that do not have a quantum mechanical origin. However, the empirical reactive force fields are substantially faster than DFTB—allowing access to even larger system sizes and time scales that can aid researchers to better illustrate and understand the ferroelectric phase transition in 2D HOIP.

Examples of contemporary reactive force field methods include MEAM105 AIREBO,106 COMB107–110 and ReaxFF.100,101 Of these methods, ReaxFF, which combines a bond order concept111 with the EEM polarizable charge transfer concept112 has arguably demonstrated the largest transferability—showing applications to a wide range of organic and inorganic materials and their interfaces (Fig. 6). Furthermore, ReaxFF has been successfully used with mixed Monte Carlo/MD schemes—like force biased Monte Carlo (fMC)113–115—extending effective time scales to the microsecond range necessary for simulating slow chemical aging and oxidation steps at hybrid interfaces.

To this date, no ReaxFF description for HOIPs has been reported yet. However, ReaxFF and other reactive force field concepts have been successfully applied to various metal-organic frameworks116–119 and to ferroelectric oxides.120–122 Furthermore, non-reactive neural network force fields have been reported for organic perovskites.123 This indicates that organic perovskites and their interfaces with other materials are a viable target for the ReaxFF force field class. One major challenge is the large variety in the organic perovskite material space. This, by itself, is already a major parametrization target for a parameter-heavy method like ReaxFF. Combining this with an organic perovskite/ interface description makes this an even harder target. Here, we believe that the introduction of machine learning (ML) methods into the ReaxFF parameterization process can add significant speedup and quality to this parameterization process. ML methods can identify parameter correlations and impose periodic-table based trends on top of the ReaxFF parameters—and can also parallelize the parameterization process. We recently demonstrated that an ML-based training concept, combined with a simple brute-force optimization, can already reduce the ReaxFF training time by a factor ten.124 Further improvements in the ML optimization tools can further streamline this process and the physical consistency of the resulting parameter sets, making development and application of complex reactive force field concepts, like ReaxFF, viable for HOIPs and their interfaces with other materials.

VII. CONCLUSION

Since the early work on HOIP in 1978125 and demonstration of HOIP as photovoltaic light absorbers in 2009,126 this material family has attracted significant research interest. The compositional and structural tunability gives rise to multiple interesting physical properties including ferroelectricity. Especially, the 2D and quasi-2D structured HOIPs exhibit promising ferroelectric properties including the superior Curie temperature and considerably high spontaneous polarization. In this perspective, we have briefly introduced the structure of 2D and quasi-2D HOIPs and presented examples of work on 2D HOIP ferroelectrics and their corresponding ferroelectric phase transition and discussed the potential of these materials in future device application. Next, we discussed the current challenges for this type of ferroelectrics and provided the research directions that could be beneficial for future studies. Finally, we briefly discussed the promising potential of machine learning (ML) on the new material discovery, and the possibility to model the property of hybrid organic/inorganic material interface via ML-enhanced atomistic simulation, which could aid the design and development of novel 2D HOIP ferroelectric systems. Several potential directions for future research are identified here:

(1) Molecular design A-site: The large A-site space in 2D HOIPs offers a large molecular design capacity which leads to excellent tunability of ferroelectric nature. Understanding the correlation of molecular dipole as well as their orientational alignment of A-site spacers to the macroscopic polarization is critical in the further development of high-performance ferroelectric 2D HOIPs. Prior attempts have introduced several strategies to enlarge the asymmetry of A-site molecules such as H/F substitution and chirality incorporation. Further enlarging
the molecular dipole moment of A-site molecules may rely on other new mechanisms.

(2) Multiferroics: Multiferroics are materials that exhibit more than one ferroic properties. These materials have potential in various applications including actuators, sensors, and memory devices. 2D HOIPs provide the opportunity for incorporating an additional property such as a magnetic feature by hypothetically replacing the B-site metals with spin-asymmetric elements such as Fe and Mn. In fact, 2D Heisenberg HOIP ferromagnets have been realized but exhibit the magnetic response at very low temperatures. Considering the excellent compositional tunability of 2D HOIPs, they can be a promising platform to develop novel multiferroics materials.

(3) Lead-free: Developing lead-free ferroelectrics is essential due to the concerns of the toxic Pb element. Future development of lead-free 2D HOIP ferroelectric materials could help release the environmental burden of Pb-based ferroelectrics and enables environmental-friendly devices. Similar to the B-site doping strategy for multiferroics, replacing the lead element by other metal or even organic moiety could be possible solutions. In this direction, mining the new materials with the help of an intelligent machine learning algorithm is of great importance.

(4) Stability: Although the 2D HOIPs are more durable than their 3D counterparts, their environmental stability is still intrinsically inferior compared to the traditional oxide perovskites. Further improving the material stability of these materials by molecular design is of high interest. For instance, H/F substitution on A-site organic spacers can not only enlarge dipole moment but also enhance moisture stability due to the hydrophobicity of F atoms. Modifying the crystal packing by tuning the steric arrangement of A-site spacers to retard environmental stimuli such as moisture and heat is another route to further enhance the durability of 2D HOIP ferroelectrics. For this purpose, the chemical aspects such as bonding strength, phase stability, and ionic activation energy are important considerations.

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DATA AVAILABILITY

The data that support the findings of this study are available within the article.

REFERENCES


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