

RESEARCH ARTICLE

Enhanced Bulk Photovoltaics in Polar 2D Hybrid Perovskite via Pressure-Strengthened Organic-Inorganic Interactions

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ABSTRACT

The development of bulk photovoltaic effect (BPVE) in polar two-dimensional (2D) organic-inorganic hybrid perovskites (OIHPs) is currently impeded by an insufficient understanding of the microscopic mechanisms linking organic-inorganic interactions to photovoltaic output. Here, we utilize pressure as an effective thermodynamic probe to decouple these interactions in BDA(EA)₂Pb₃Br₁₀ (BDA is 1,4-butanediammonium, EA is ethylammonium), achieving the first pressure-engineered synergistic enhancement of short-circuit current (I_{SC}) and open-circuit voltage (V_{OC}). Comprehensive in situ characterizations reveal that pressure-strengthened N—H···Br hydrogen bonding induces cooperative organic cation ordering and inorganic octahedral distortion. This structural synergy amplifies spontaneous polarization, resulting in a 530-fold increase in BPVE efficiency to 0.18% at 1.5 GPa, a value surpassing all previously reported OIHPs. Moreover, the enhanced organic-inorganic interaction stabilizes the ferroelectric state, elevating the Curie temperature to 433 K. These results elucidate the microscopic mechanism underlying BPVE enhancement in 2D OIHP ferroelectrics and establish pressure modulation as an effective strategy for mechanistic studies and photovoltaic optimization.

1 | Introduction

The bulk photovoltaic effect (BPVE), which enables the generation of photocurrent and photovoltage in non-centrosymmetric materials without the need for p-n junctions, has attracted increasing attention as a promising paradigm for next-generation photovoltaic and self-powered optoelectronic devices [1–5]. Unlike conventional photovoltaic mechanisms that rely on heterostructure design, BPVE originates from intrinsic symmetry

breaking [6, 7]. Polar 2D organic-inorganic hybrid perovskites (OIHPs) have emerged as compelling BPVE-active materials owing to their unique structural and compositional tunability [8–10]. In these materials, the coexistence of polar arrangement of organic cations and distorted inorganic frameworks gives rise to spontaneous polarization, enabling BPVE responses [11–13]. However, the BPVE performance of 2D OIHP ferroelectrics remains far from practical requirements [14, 15]. This challenge is largely due to the lack of a clear microscopic picture

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linking organic-inorganic interactions, structural polarization, and BPVE output, which has so far prevented the establishment of rational optimization strategies for simultaneously enhancing short-circuit current (I_{SC}) and open-circuit voltage (V_{OC}) [16, 17].

To overcome these limitations, major efforts have been devoted to chemical composition engineering, such as tailoring organic cation dipoles or substituting inorganic ions [18–20]. Whereas, these chemical methods are intrinsically limited by their discrete nature; they require the synthesis of a new sample for each state, which precludes continuous modulation of the electronic structure [21, 22]. In addition, chemical substitution precludes in situ characterization of the continuous structure-property evolution, thereby hindering the elucidation of underlying mechanisms [23, 24]. As a thermodynamic parameter, pressure offers a powerful means to effectively and continuously tune interatomic distances, lattice distortions, and organic-inorganic interactions without altering chemical composition [25–29]. While pressure has been shown to effectively regulate crystalline structures and electronic properties in various functional materials, its impact on the hybrid structure and BPVE performance of 2D OIHP ferroelectrics has remained largely unexplored.

In this work, $BDA(EA)_2Pb_3Br_{10}$ was selected as the model system owing to its non-centrosymmetric ferroelectric structure, a prerequisite for the BPVE, and its Dion-Jacobson (DJ) framework, which facilitates strong organic-inorganic interactions. This combination provides a suitable platform for examining how pressure-modulated organic-inorganic interactions affect the BPVE response. We report the first demonstration of pressure-engineered enhancement of the BPVE in a polar 2D OIHP, achieving a synergistic increase in both I_{SC} and V_{OC} . By precisely tuning organic-inorganic interactions, we realize a 45.8-fold enhancement in I_{SC} and an 11.5-fold increase in V_{OC} at 1.5 GPa, leading to a remarkable 530-fold improvement in BPVE efficiency of 0.18%, a value superior to reported OIHPs, including $(EA)_4Pb_3Cl_{10}$ [30], $(n\text{-PA})_2(EA)_2Pb_3I_{10}$ [31], $(ALA)_2(EA)_2Pb_3Br_{10}$ [32], and $(EA)_2(MA)_2Pb_3Br_{10}$ [33] with $n\text{-PA}$ = n -pentylammonium, ALA = allylammonium, MA = methylammonium. Comprehensive in situ high-pressure structural and spectroscopic characterizations reveal that pressure-enhanced $N\cdots Br$ hydrogen bonding promotes orientational ordering of BDA^{2+} cations and increases distortions of $PbBr_6$ octahedra. These cooperative structural evolutions strengthen the overall polarization, thereby boosting the BPVE response. Moreover, enhanced organic-inorganic interaction effectively stabilizes the ferroelectric state, elevating the Curie temperature (T_C) from 373 K at ambient conditions to 433 K at 1.9 GPa. This work establishes pressure as an effective route for optimizing photovoltaic properties in 2D OIHPs and provides fundamental insights into the interplay between organic-inorganic interactions, ferroelectricity, and BPVE performance.

2 | Results and Discussion

2.1 | Pressure-Enhanced BPVE in $BDA(EA)_2Pb_3Br_{10}$

To investigate the pressure dependence of BPVE in $BDA(EA)_2Pb_3Br_{10}$, an electrical device with a two-electrode

configuration was fabricated in a diamond anvil cell (DAC) chamber (Figure 1a). The crystal was pre-polarized by applying a -5 V bias for 120 s, after which photocurrent measurements were carried out at zero bias under 405 nm illumination. As shown in Figure 1b, the current-voltage (I - V) curves do not pass through the origin, a characteristic feature of the BPVE [34]. This behavior originates from the spontaneous polarization in polar 2D OIHPs, which gives rise to I_{SC} and V_{OC} [35]. The reproducible on-off photoresponses under 405 nm illumination at zero bias demonstrate that the BPVE in $BDA(EA)_2Pb_3Br_{10}$ remains robust under compression (Figure 1c). With increasing pressure up to 1.5 GPa, I_{SC} rises from 9.6 pA to 0.44 nA and V_{OC} from 0.06 to 0.69 V, corresponding to 45.8-fold and 11.5-fold enhancements relative to the initial state, respectively (Figure 1d). As a result, the BPVE efficiency of $BDA(EA)_2Pb_3Br_{10}$ reaches 0.18% at 1.5 GPa, which is approximately 530 times higher than the initial state, and then decreases at higher pressures (Figure 1e; Figure S1). The corresponding I - V curves collected during decompression are shown in Figure S2, revealing that the pressure-dependent photovoltaic response is largely reversible upon pressure release. Importantly, the $BDA(EA)_2Pb_3Br_{10}$ achieves a BPVE efficiency of 0.18% under pressure that exceeds that of all polar 2D OIHPs and most reported traditional ferroelectric materials, including $9.8 \times 10^{-3}\%$ for $(EA)_4Pb_3Cl_{10}$ [30], $9.7 \times 10^{-5}\%$ for $(n\text{-PA})_2(EA)_2Pb_3I_{10}$ [31], $2.6 \times 10^{-4}\%$ for $(ALA)_2(EA)_2Pb_3Br_{10}$ [32], $1.0 \times 10^{-3}\%$ for $(EA)_2(MA)_2Pb_3Br_{10}$ [33], and most reported traditional ferroelectric materials (e.g., $BiFeO_3$ (BFO), $1.3 \times 10^{-2}\%$; $BaTiO_3$ (BTO), $2.1 \times 10^{-4}\%$; $[KNbO_3]_{1-x}[BaNi_{1/2}Nb_{1/2}O_{3-\delta}]_x$ (KBNN), $3.5 \times 10^{-4}\%$) (Figure 1f and Table S1) [36–38]. In addition, we evaluated the factors that may affect BPVE efficiency under pressure, and the detailed discussion is provided in Tables S2 and S3; Figure S3; Figure S4.

We further performed pressure-dependent BPVE measurements on several other representative polar 2D OIHPs (Figures S5–S7 and Table S4). The results reveal a more pronounced BPVE enhancement in DJ perovskite $BDA(EA)_2Pb_3Br_{10}$ than in Ruddlesden-Popper (RP) perovskites (530-fold for DJ $BDA(EA)_2Pb_3Br_{10}$ vs. 2.9- to 10.2-fold for several RP perovskites). This difference could be associated with stronger hydrogen-bond-mediated organic-inorganic coupling in DJ structures, which can facilitate the transfer of compression to the polar inorganic framework. In contrast, the softer van der Waals interlayers in RP structures can accommodate the applied compression, thereby limiting its transfer to the inorganic framework and reducing the BPVE enhancement.

Having established the remarkable enhancement of the BPVE under pressure, we then investigated its polarization dependence to uncover the underlying mechanism. At ambient conditions, $BDA(EA)_2Pb_3Br_{10}$ exhibits a clear photovoltaic response under uniform illumination, as evidenced by its I - V curves (Figure S8). By applying poling voltages in opposite directions, both V_{OC} and I_{SC} can be effectively reversed, indicating that the BPVE is closely associated with the ferroelectric polarization of the material. We applied successive poling pulses under pressure to explore how ferroelectric polarization regulates the photovoltaic response of $BDA(EA)_2Pb_3Br_{10}$. As shown in Figure S9, at 0.5 GPa, a preset pulse of -5 V was first applied to initialize the polarization in the downward direction, giving a V_{OC} of about -0.36 V and an

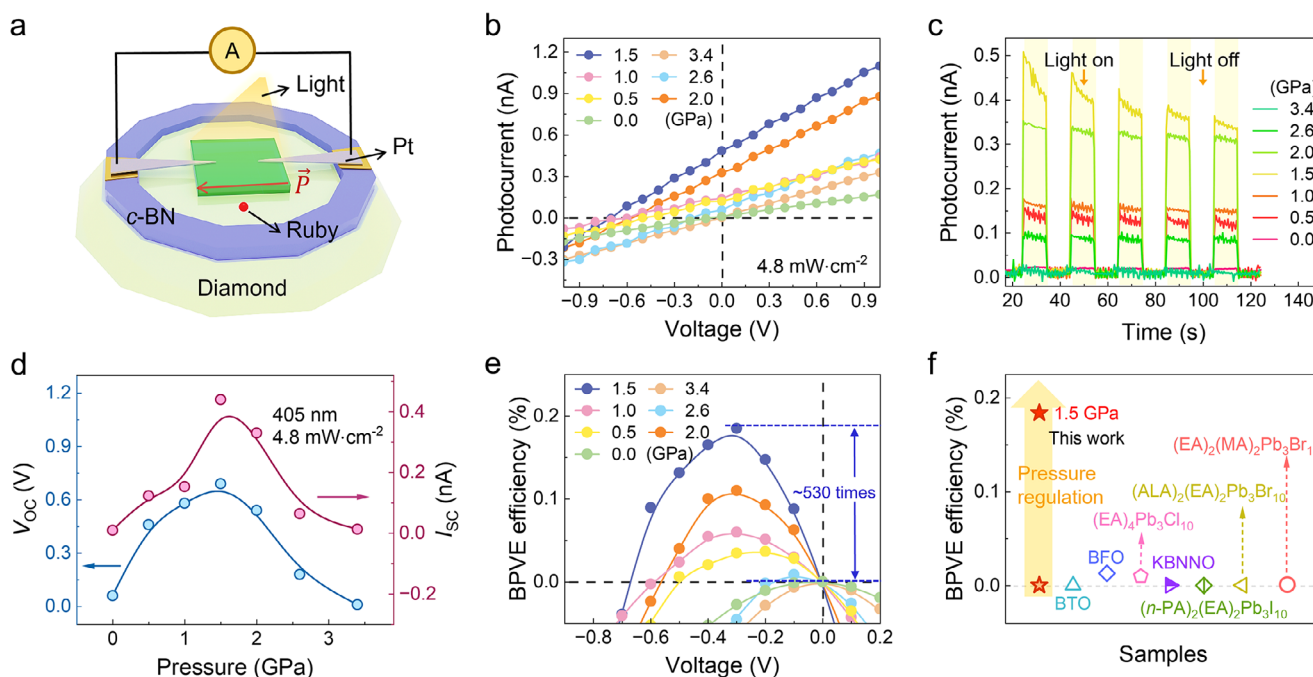


FIGURE 1 | Pressure-dependent BPVE characterization of BDA(EA)₂Pb₃Br₁₀ under 405 nm illumination. (a) Schematic of the two-electrode device assembled inside a DAC for BPVE measurements. (b) *I*-*V* curves of BDA(EA)₂Pb₃Br₁₀ under different pressures. (c) Reproducible photocurrents were recorded during on-off light cycles at zero bias under compression. (d) *V*_{OC} (blue) and *I*_{SC} (red) as a function of pressure. (e) BPVE efficiency as a function of voltages at different pressures. (f) Comparing the BPVE efficiency of reported ferroelectric materials. Data for other materials were taken from references (BTO [37], BFO [36], KBNNO [38], (EA)₄Pb₃Cl₁₀ [30], (n-PA)₂(EA)₂Pb₃I₁₀ [31], (ALA)₂(EA)₂Pb₃Br₁₀ [32], (EA)₂(MA)₂Pb₃Br₁₀ [33]). The connecting lines are guides to the eye and do not represent fits to the data.

*I*_{SC} of 0.09 nA. As the positive poling voltage gradually increased, the *V*_{OC} evolved from negative to positive values, while the *I*_{SC} shifted toward the negative direction. This progressive shift indicates a gradual evolution of the macroscopic polarization state and the corresponding modulation of the internal electric field. Conversely, when a reverse voltage was applied to the sample previously poled at 30 V, both *V*_{OC} and *I*_{SC} gradually reversed. Similar behavior was also observed at 1.0 and 1.5 GPa (Figure S10; Figure S11), demonstrating that BDA(EA)₂Pb₃Br₁₀ retains a polarization-switchable photovoltaic response under pressure, which underlies its effectively tunable photoresponse. In addition, the evolution of *V*_{OC} and *I*_{SC} as a function of poling voltage exhibits clear hysteresis-like characteristics (Figure S12), revealing the intrinsic correlation between the BPVE response and the polarization state. We also examined the stability of *I*_{SC} under continuous illumination (Figure S13). The stable *I*_{SC} output indicates good operational stability and nonvolatile behavior under pressure. These results further confirm that the BPVE in BDA(EA)₂Pb₃Br₁₀ is effectively regulated by ferroelectric polarization.

To probe the light-intensity-dependent behavior of the BPVE, we further measured the *I*-*V* curves and on-off photoresponses at 2.0 GPa under different incident light power densities (Figure S14; Figure S15). With increasing light power densities, the *I*_{SC} increases while the *V*_{OC} remains nearly constant (Figure S16). Moreover, the responsivity (*R*) and specific detectivity (*D*^{*}) of BDA(EA)₂Pb₃Br₁₀ were evaluated under different pressures (Figure S17 and Table S5). Under 405 nm illumination with a light intensity of 4.8

mW·cm⁻², both *R* and *D*^{*} reach their maximum values at 1.5 GPa, attaining 1.02×10^{-2} A·W⁻¹ and 3.38×10^{10} Jones, respectively. These values surpass those of several reported self-powered photodetectors based on 2D OIHPs, including (*i*-BA)₂MAPb₂Cl₇ [39], (EA)₂(MA)₂Pb₃Br₁₀ [33], (EA)₄Pb₃Cl₁₀ [30], and the (4-AMP)(MA)₂Pb₃Br₁₀/MAPbBr₃ heterostructure [40], where *i*-BA and 4-AMP denote isobutylammonium and 4-(aminomethyl)piperidinium, respectively. In addition, both *R* and *D*^{*} gradually decrease with increasing light intensity, a behavior commonly observed in hybrid perovskite-based photodetectors (Figure S18) [41].

2.2 | Pressure-Induced Octahedral Distortion and Structural Phase Transition

The pressure-induced structural evolution correlates closely with the enhanced BPVE observed in BDA(EA)₂Pb₃Br₁₀. To clarify this relationship, in situ synchrotron powder X-ray diffraction (XRD) measurements were carried out from ambient conditions to 2.8 GPa (Figure 2b; Figure S19). At ambient conditions, BDA(EA)₂Pb₃Br₁₀ crystallizes in a 2D perovskite structure with polar space group *Pnm*2₁ (Phase I). The structure features adjacent inorganic layers (*L*₁ and *L*₃) bridged by hydrogen bonds at both termini of the BDA²⁺ cations, while EA⁺ cations occupy the cavities within the inorganic octahedral framework (Figure 2a). These organic-inorganic hydrogen-bonding interactions help stabilize the ambient-pressure polar structure by coupling organic-cation orientations to distortions of the inorganic PbBr₆ octahedra [42–44]. As pressure increases, all Bragg diffraction peaks shift to

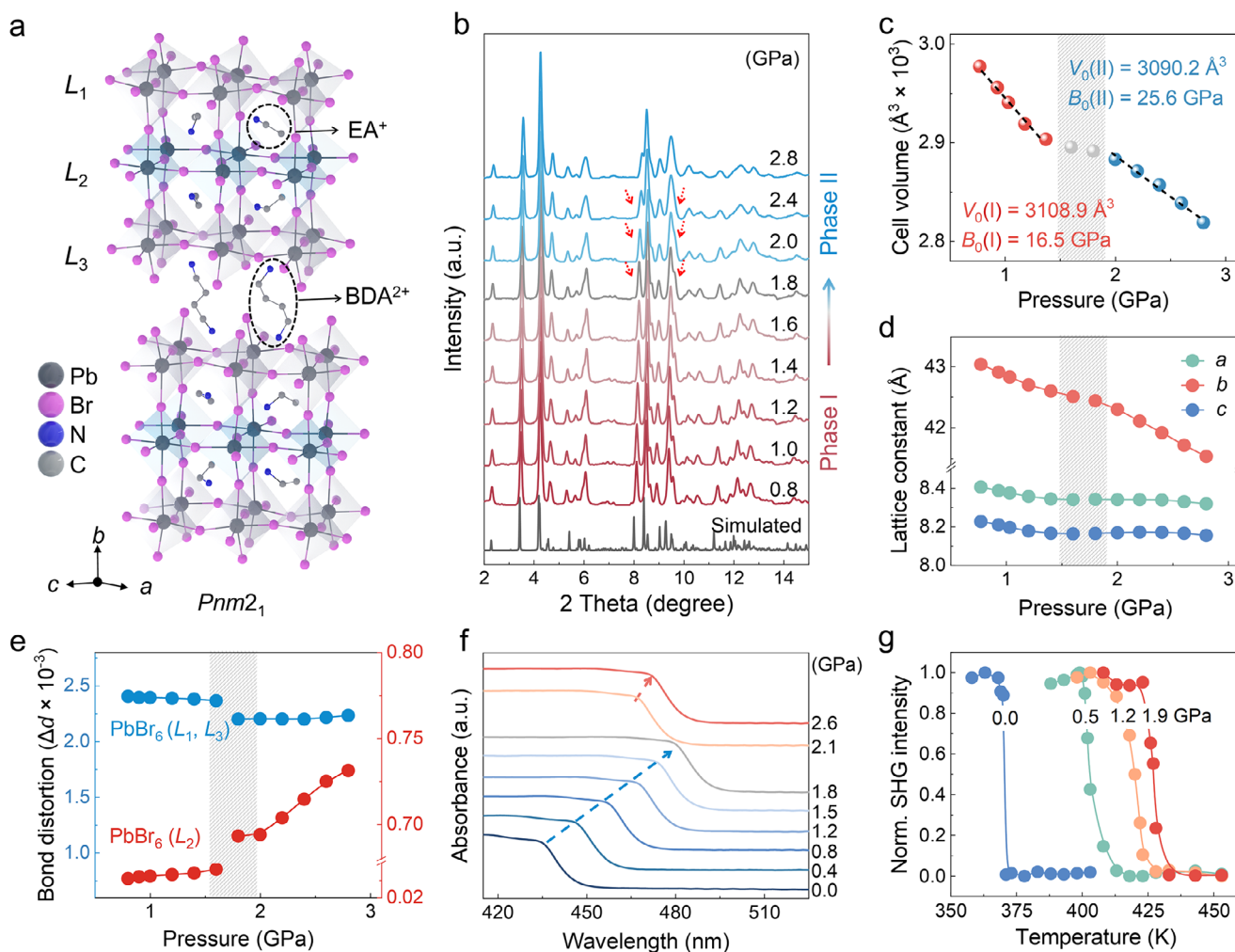


FIGURE 2 | Evolutions of crystal structure and optical properties of BDA(EA)₂Pb₃Br₁₀ during compression. (a) Crystal structure of BDA(EA)₂Pb₃Br₁₀ at ambient conditions, with hydrogen atoms omitted for clarity. (b) In situ synchrotron powder XRD patterns of BDA(EA)₂Pb₃Br₁₀ at selected pressures. (c) Unit cell volumes as a function of pressure. The values of bulk modulus B_0 were determined to be 16.5 and 25.6 GPa in phases I and II, respectively. (d) Variations of lattice constants during compression. (e) Pressure-dependent variations of the bond distortion index Δd , where Δd describes the degree of octahedral distortion. (f) UV-Vis absorption spectra at selected pressures. (g) Normalized SHG intensities as a function of temperature at selected pressures, which determine the Curie temperature.

higher two-theta angles due to the lattice contraction. At 1.8 GPa, several diffraction peaks merge, indicating a pressure-driven structural phase transition. Rietveld refinements further confirm the transition from the orthorhombic $Pnm2_1$ phase (Phase I) below 1.8 GPa to the orthorhombic $Ama2$ phase (Phase II) at 1.8 GPa and above, with R_p values of 0.016–0.018 and R_{wp} values of 0.029–0.037 (Figure S19). The corresponding refined lattice constants and unit-cell volumes are summarized in Table S6.

The pressure dependence of the unit-cell volumes (Figure 2c and Table S6) was analyzed using the second-order Birch-Murnaghan equation of state, yielding bulk moduli $B_0 = 16.5$ GPa for Phase I and $B_0 = 25.6$ GPa for Phase II (Figure 2c). Both phases exhibit pronounced anisotropic compressibility, with the b axis contracting faster than the a and c axes (Figure 2d). This anisotropy indicates pressure-enhanced interlayer organic-inorganic interactions that directly influence the octahedral distortion of PbBr₆ units. To quantify the octahedral distortion,

we employ the bond distortion index Δd [45, 46]:

$$\Delta d = \frac{1}{6} \sum_{i=1}^6 \left(\frac{|d_i - \bar{d}|}{\bar{d}} \right)^2 \quad (1)$$

where d_i and $\bar{d}$ are the individual and average Pb–Br bond lengths, respectively. Below the phase transition, Δd increases progressively with pressure, indicating enhanced octahedral distortion. This structural evolution strengthens polarization, thereby contributing to the enhancement of BPVE performance (Figure 2e).

Pressure-induced structural variation also profoundly affects the optical properties of BDA(EA)₂Pb₃Br₁₀. We traced the evolution of the bandgap under high pressure using in situ UV-Vis absorption spectroscopy (Figure 2f). Upon compression, the bandgap undergoes a gradual redshift, but abruptly increases when pressure

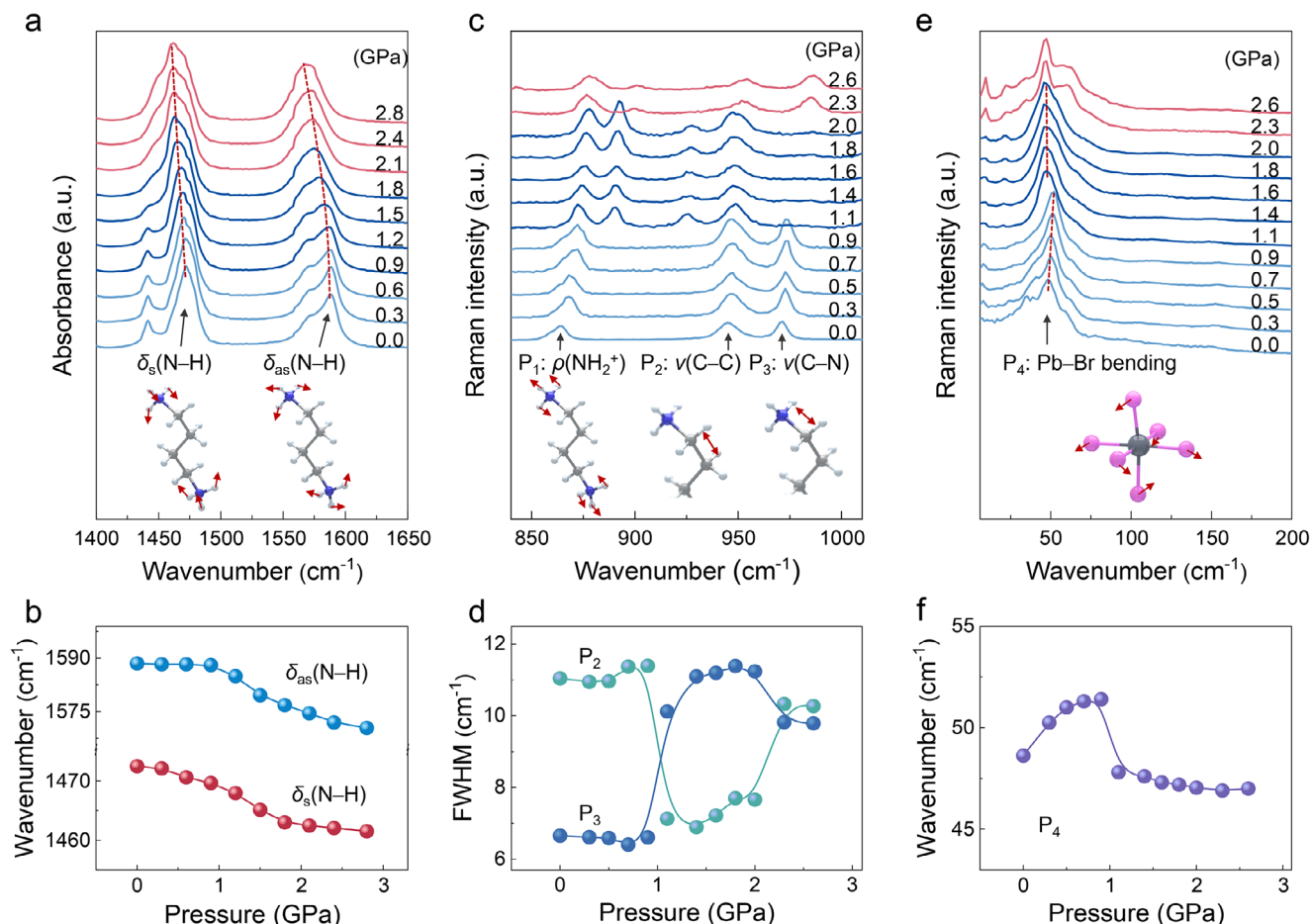


FIGURE 3 | Infrared and Raman spectroscopic characterizations of BDA(EA)₂Pb₃Br₁₀ under pressure. (a) Pressure-dependent infrared spectra of BDA(EA)₂Pb₃Br₁₀ in the 1400–1650 cm⁻¹ range. Inset graphs illustrate the corresponding vibration modes. (b) Pressure dependence of infrared wavenumbers for the δ_s(N-H) and δ_{as}(N-H) vibrational modes. (c) Pressure-dependent Raman spectra in the 840–1010 cm⁻¹ range, where P₁, P₂, and P₃ modes correspond to the ρ(NH₂⁺) rocking vibration, the ν(C–C) stretching vibration, and the ν(C–N) stretching vibration of BDA²⁺ cations. (d) FWHM of the P₂ and P₃ modes as a function of pressure. (e) Pressure-dependent Raman spectra in the 10–200 cm⁻¹ range, where the P₄ mode corresponds to the Pb–Br bending vibration of the PbBr₆ octahedra. (f) The evolution of the P₄ mode as a function of pressure.

exceeds 1.8 GPa, consistent with the occurrence of a structural phase transition (Figures S20 and S21). Furthermore, strengthened organic-inorganic interactions under pressure are expected to raise the T_c . To verify this, temperature-dependent second harmonic generation (SHG) measurements were performed on BDA(EA)₂Pb₃Br₁₀ at selected pressures (Figure 2g). During compression, T_c increases from 373 K at ambient conditions to 433 K at 1.9 GPa. This substantial increase in T_c not only suggests that strengthened organic-inorganic interactions effectively stabilize the ferroelectric state but also highlights the crucial role of pressure in enhancing the BPVE performance by promoting the orientational ordering of BDA²⁺ cations.

2.3 | Structure Evolution and Organic Cation Vibration Under Pressure

To further elucidate the influence of organic-inorganic interactions on interlayer cation ordering in BDA(EA)₂Pb₃Br₁₀, we performed in situ infrared and Raman spectroscopy measurements (Figure 3). As illustrated in Figure S22, hydrogen bonding between interlayer BDA²⁺ cations and the inorganic framework

stabilizes an ordered polar structure. Under compression, the δ_s(N–H) and δ_{as}(N–H) modes in the infrared spectra exhibit pronounced redshifts and broadening, indicating N–H bond elongation (Figure 3a,b), which is a signature of strengthened organic-inorganic interactions [47]. Correspondingly, the δ_s(NH₂⁺) vibrational mode (P₅) and δ_{as}(NH₂⁺) vibrational modes (P₆, P₇) in the Raman spectra also redshift with pressure (Figure S23 and Table S7), corroborating the enhancement of organic-inorganic interactions.

These intensified organic-inorganic interactions synergistically regulate both organic cation orientation and inorganic octahedral distortion. Upon compression, the ρ(NH₂⁺) rocking mode (P₁) splits into two sharp bands, signaling an ordered reorientation of BDA²⁺ cations to accommodate lattice contraction (Figure 3c). Concurrently, the full width at half maximum (FWHM) of the ν(C–C) stretching mode (P₂) narrows sharply, while that of the ν(C–N) mode (P₃) broadens progressively, reflecting enhanced spatial confinement of the BDA²⁺ cations (Figure 3d). Notably, the Pb–Br bending vibration mode (P₄) of the PbBr₆ octahedra redshifts above 1.1 GPa (Figure 3e,f; Figure S24), suggesting interaction between inorganic lattice vibrations and BDA²⁺

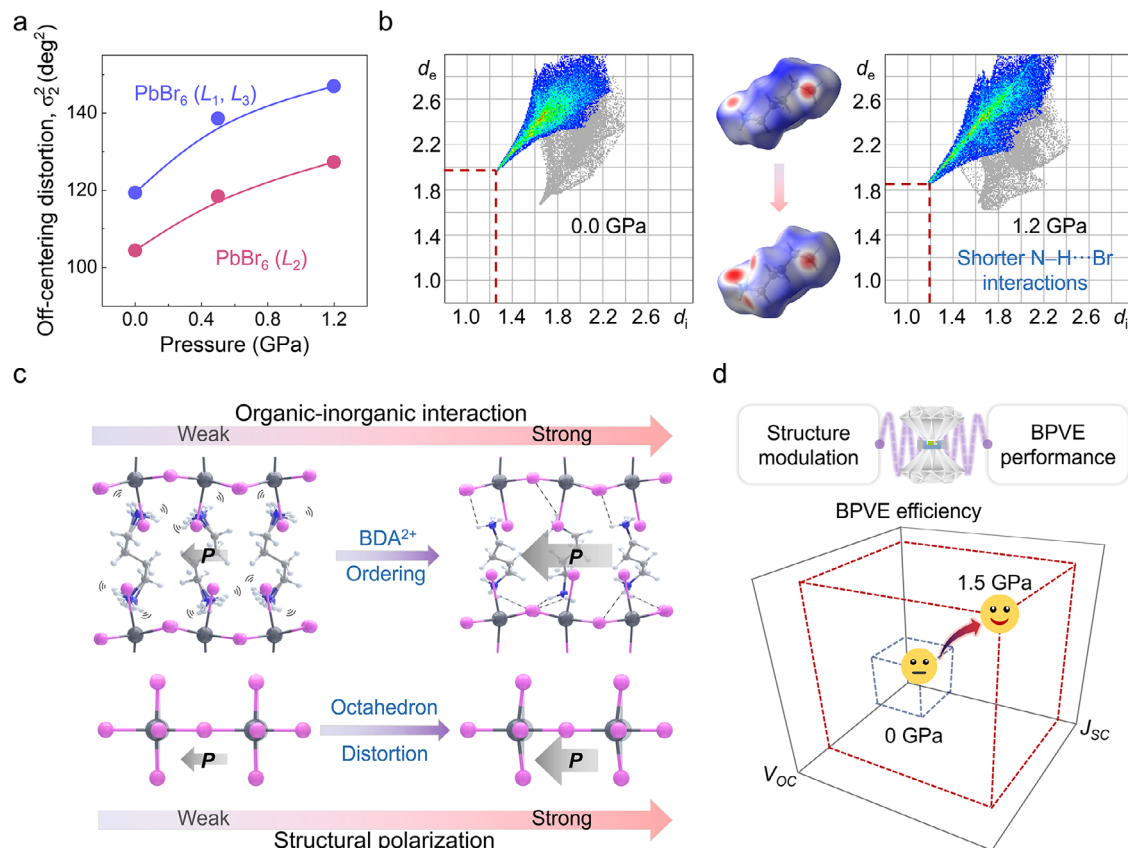


FIGURE 4 | Mechanism of pressure-enhanced BPVE in BDA(EA)₂Pb₃Br₁₀. (a) Pressure dependence of the Pb off-centering distortion σ_2^2 for the L_1/L_3 and L_2 PbBr₆ octahedra, derived from single-crystal XRD refinements. (b) 2D fingerprint plots and Hirshfeld surfaces at 0 and 1.2 GPa. (c) Enhanced organic-inorganic interactions under pressure promote the orientational ordering of BDA²⁺ cations and increase distortions of the PbBr₆ octahedra, thus enhancing the structural polarization. (d) Schematic illustration of BPVE enhancement through pressure-tuned organic-inorganic interactions.

cation motions. This interaction promotes octahedral distortion. Collectively, these findings demonstrate that the strengthening of hydrogen bonding facilitates the orientational ordering of BDA²⁺ cations and increases PbBr₆ octahedral distortions, both of which are critical for enhancing the BPVE performance of BDA(EA)₂Pb₃Br₁₀. Upon further compression into Phase II, the Raman peaks associated with the organic cations (e.g., P₁, P₂, and P₃) exhibit pronounced broadening, indicating a disruption in their orientational order. Since the BPVE is closely related to this cooperative structural ordering, such structural disorder weakens spontaneous polarization and leads to the observed decline in BPVE efficiency.

2.4 | Mechanism of Pressure-Enhanced BPVE in BDA(EA)₂Pb₃Br₁₀

We further employed high-pressure single-crystal XRD measurements within Phase I to directly verify the local structural evolution. We quantified the local polar distortion of the PbBr₆ octahedra using the *B*-cation off-centering distortion σ_2^2 , defined as [17, 48]:

$$\sigma_2^2 = \frac{1}{2} \sum_{i=1}^3 (\beta_i - 180^\circ)^2 \quad (2)$$

where β_i represents one of the three trans Br—Pb—Br bond angles in each PbBr₆ octahedron. The σ_2^2 values increase continuously with pressure for both the octahedra, indicating enhanced Pb off-centering and increased octahedral distortion (Figure 4a; Tables S8 and S9). Furthermore, single-crystal structural analysis reveals a pressure-induced compression and reorganization of the N—H...Br hydrogen-bonding network. The average N...Br distance decreases from 3.338 Å at ambient pressure to 3.244 Å at 1.2 GPa, while the shortest N...Br distance decreases from 3.322 to 3.072 Å and the number of short contacts increases from 2 to 4 (Figure 4b and Table S10). These changes reveal a more compact and reorganized hydrogen-bonding network, indicating the reorientation of the BDA²⁺ cations and enhanced organic-inorganic coupling under pressure. Consistently, Hirshfeld surface and 2D fingerprint analyses show more pronounced H...Br contact regions and a shift toward smaller d_i and d_e values under pressure (Figure 4b), further supporting a more compact N—H...Br contact network.

Building upon the knowledge obtained from the above high-pressure experiments, we can optimize the BPVE performance by controlling organic-inorganic interactions. The strengthened organic-inorganic interactions drive two key structural modifications: (i) enhanced orientational ordering of the BDA²⁺ cations and (ii) increased distortion and off-centering of the PbBr₆ octahedra. These concurrent changes in molecular orientation and octahedral geometry cooperatively enhance the structural

polarization in $\text{BDA}(\text{EA})_2\text{Pb}_3\text{Br}_{10}$. The strengthened spontaneous polarization promotes more efficient separation and transport of photogenerated charge carriers, leading to substantial increases in both V_{OC} and I_{SC} . Consequently, the BPVE performance of pressurized $\text{BDA}(\text{EA})_2\text{Pb}_3\text{Br}_{10}$ is markedly enhanced, as illustrated in Figure 4c,d.

3 | Conclusion

In summary, this work establishes pressure engineering as a powerful paradigm for optimizing the BPVE in 2D OIHP ferroelectrics. Taking $\text{BDA}(\text{EA})_2\text{Pb}_3\text{Br}_{10}$ as a model system, we achieved a synergistic enhancement of I_{SC} (45.8-fold) and V_{OC} (11.5-fold) through the precise modulation of organic-inorganic interactions. The resulting BPVE efficiency of 0.18% at 1.5 GPa represents a benchmark value among reported OIHPs. Furthermore, the reinforced organic-inorganic interaction effectively stabilizes the ferroelectric state, elevating the T_c to 433 K. Comprehensive in situ high-pressure structural and spectroscopic measurements reveal that the strengthening of N—H...Br hydrogen bonds under compression promotes the orientational ordering of BDA^{2+} cations and amplifies the distortion of PbBr_6 octahedra. These cooperative structural evolutions significantly enhance spontaneous polarization and BPVE performance. These findings not only elucidate the microscopic mechanism governing BPVE enhancement in 2D OIHPs but also establish a new degree of freedom for rationalizing the design of high-performance optoelectronics.

Author Contributions

Shiyu Fang: writing – original draft, investigation, data curation, conceptualization. **Kejun Bu:** writing – review and editing, funding acquisition, data curation. **Huang Ye:** investigation. **Lijun Xu:** investigation. **Dong Wang:** investigation. **Songhao Guo:** funding acquisition, writing – review and editing. **Tonghuan Fu:** data curation, writing – review and editing. **Yuhong Mao:** data curation, writing – review and editing. **Yuchen Ye:** investigation. **Liwei Tang:** investigation. **Philip Dalladay-simpson:** funding acquisition, writing – review and editing, resources. **Federico Aiace Gorelli:** writing – review and editing, resources. **Yiqiang Zhan:** writing – review and editing. **Yang Ding:** investigation, funding acquisition. **Zhihua Sun:** funding acquisition, writing – review and editing. **Junhua Luo:** writing – review and editing. **Xujie Lü:** funding acquisition, conceptualization, project administration, resources, supervision.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data supporting the key findings of this study are available within the article and its [Supporting Information](#) files.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

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