Strain in perovskite solar cells: origins, impacts, and regulation

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ABSTRACT

Metal halide perovskite solar cells (PSCs) have seen an extremely rapid rise in power conversion efficiencies in the past few years. However, the commercialization of this class of emerging materials still faces serious challenges, one of which is the instability against external stimuli such as moisture, heat, and irradiation. Much focus has deservedly been placed on understanding the different origins of intrinsic instability and thereby enhancing their stability. Among these, tensile strain in perovskite films is an important source of instability that cannot be overcome using conventionally extrinsic stabilization approaches such as encapsulation. Here we review recent progress in understanding of the origin of strain in perovskites as well as its corresponding characterization methods, and their impacts on the physical properties of perovskites and the performance of PSCs including efficiency and stability. We then summarize the latest advances in strain-regulation strategies that improve the intrinsic stability of perovskites and photovoltaic devices. Finally, we provide a perspective on how to make further progress in stable and high-efficiency PSCs via strain engineering.

Keywords: perovskite, stress, strain, stability, solar cells

INTRODUCTION

The power conversion efficiencies (PCEs) of perovskite solar cells (PSCs) have improved rapidly from 3.8% to a certified 25.2% for single junction devices and approaching to 30% for perovskite-based tandem devices in the past few years [1-6]. Such excellent performance can be mainly attributed to their long carrier diffusion lengths and low trap densities arising from their defect-tolerant properties [7-10]. However, the instability of PSCs remains as the largest barrier toward their commercialization [11-14]. Lead halide perovskites have been reported to be sensitive to many external stimuli, such as moisture, oxygen, heat, and ultraviolet light [15,16]. Recent
efforts have demonstrated progress in enhancing the stability of PSCs extrinsically by introducing hydrophobic coating, replacing reactive metal electrodes with noncorrosive carbon or transparent conducting oxides, and encapsulation techniques [17-20]; these approaches straightforwardly protect perovskite devices from the ambient environment. Once the degradation of PSCs induced by external stimuli is suppressed, the intrinsic instability of perovskite material itself—which cannot be solved using extrinsic protection methods—would impose the limitation on the stability of whole devices [21].

Perovskites especially the organic-inorganic hybrid perovskites possess a flexible crystal structure owing to their soft lattice. This enables the manipulation of structural and optoelectronic properties of perovskites through strain engineering [22-25]. For instance, by applying appropriate hydrostatic pressure, the bandgap of CH$_3$NH$_3$PbI$_3$ becomes narrower that broadens the absorption spectrum of this material, while the carrier lifetime is prolonged, both of which are beneficial for achieving better photovoltaic performance [26-30]. When further applying the pressure as high as 60 GPa, CH$_3$NH$_3$PbI$_3$ exhibits metallic character through apparent bandgap closure. This pressure-induced semiconductor-to-metal transition demonstrates the realization of a wholly new electronic structure and transport properties in perovskites, greatly enriching the electronic diversity of perovskites [31,32]. Nevertheless, strain especially the tensile strain in perovskite has known to contribute to instability in these materials; that weakens bonds, favors the formation of defects, and lowers the activation energy for ion migration, thereby accelerating the degradation of perovskites [31,33,34]. Tensile-strain-induced intrinsic instability is now pointed to widely as a major bottleneck toward the achievement of stable PSCs [21,35].

Here we review the latest advances that correlate the strain in perovskites with their physical properties and photovoltaic performance including efficiency and stability and discuss the avenues for further progress towards high-efficiency and stable PSCs via strain regulation (Fig. 1). We begin with a discussion of two different origins of strain/stress in perovskites: (i) local lattice strain, which originates from the ionic size mismatch between the A cation and the lead halide cage size in ABX$_3$, and the local lattice mismatches due to the inhomogeneity in mixed halide perovskites; (ii) external condition-induced strain, including thermal expansion mismatch between perovskites and the contacting functional layers, and lattice mismatch between perovskites and epitaxial substrates. We then discuss the characterization of strain, and its impacts on the physical properties of perovskites (electronic structure, ion migration, and defect) and the efficiency and stability of PSCs. We follow with a summary of the recent advances in strain-regulation methods that release the tensile strain and thereby improve the intrinsic stability of perovskites and corresponding photovoltaic devices. Finally, we provide a perspective on the further strain-engineering innovations for high-efficiency and stable PSCs.

**ORIGINS OF STRAIN IN PEROVSKITES**

Since the first finding of residual strain in perovskites reported in 2015, much concern has been raised towards the strain-induced issues of PSCs especially the material and device instability [36]. Understanding the origin of strain would be therefore an essential precondition to fabricate stable PSCs [21]. In this section, we provide a brief review on the origins of strain in perovskite, excluding conventional strain-engineering methods such as
hydrostatic pressurization and bending the flexible substrates in either a concave or convex shape. There are generally two different origins of strain in perovskites: local lattice strain; external condition-induced strain.

**Local lattice strain**

Metal halide perovskites have the general formula $ABX_3$ [37,38], where $A$ is a monovalent cation, such as organic methylammonium (MA, CH$_3$NH$_3$)$^+$ [39,40] or formamidinium (FA, (NH$_2$)$_2$CH$^+$) [7,41], and inorganic cesium or rubidium ion; $B$ is a divalent metal cation, including Pb$^{2+}$, Sn$^{2+}$ or Ge$^{2+}$; and $X$ is a mixture of halogen anion (Cl, Br and I) [42-47]. The stable crystal structures of perovskites can be predicted by a reliable empirical index, Goldschmidt tolerance factor ($t$). That is calculated from the ionic radius of the atoms as follows: $t = (R_A + R_X)/[\sqrt{2}(R_B + R_X)]$, where $R_A$, $R_B$ and $R_X$ are the ionic radii of the corresponding ions. Metal halide perovskites tend to form an ideal cubic structure when $0.8<t<1$, orthorhombic structure when $t<0.8$, and hexagonal structure when $t>1$[48-50]. Besides structural stability, the size of ions also affects the stability of perovskites in ambient air [7]. Sargent *et al.* [51] revealed that mixed CsMAFA perovskites were shown better stability than FAPbI$_3$. They found that this instability of FAPbI$_3$ was attributed to the local lattice strain. This strain arises from the ionic size mismatch between FA cation and the lead iodide cage size owing to the large size of FA cation, leading to cage distortions and PbI$_6$ octahedra tilting (Fig. 2a-c). Therefore, perovskite structure with ionic size mismatch would tend to form local lattice strain.

Local lattice mismatch is another origin of local lattice strain in perovskites especially the mixed halide perovskites, despite the fact that the highest PCEs of PSCs are mostly achieved by employing these perovskites. Mixed halide perovskites have been known to suffer from materials inhomogeneity due to the composition separation into separated Br-rich and I-rich phases when exposed to heat and light. This is attributed to the substantial chemical mismatch among each component, and the unbalanced growth conditions of each component during the process of film fabrication. Recently, Chen *et al.* [35] revealed that this inhomogeneity in perovskite films perpendicular to the substrate resulted in the local lattice mismatch, then the lattice distortion of microscopic crystal structure, and consequently the local lattice strain (Fig. 2d-g), which may be a type of microstrain. Besides local lattice mismatch, local crystal misorientation can lead to local strain within perovskite grains. The grain-to-grain orientation spread further results in local strain heterogeneity within halide perovskite films (Fig. S1a). Ginger *et al.* [52] have detailed this origin of local strain through ultrasensitive electron backscatter diffraction (EBSD), which can reveal crystal orientation variations in sub-grain structure. They demonstrated the correlation between misorientation-induce local strain and misorientation angles obtained by EBSD and found that the larger misorientation angle led to higher degrees of strain (Fig. 2h).

**External condition-induced strain**

The external condition-induced strain originates from two types of mismatches: thermal expansion and lattice mismatch between the perovskite and substrate, which may be assigned to biaxial strain. Figure 3a summarizes the linear thermal expansion coefficients ($\alpha$) of lead-halide perovskites, substrates, and other functional layers including electron-transport layers (ETLs) and hole-transport layers (HTLs). Perovskites possess high values of $\alpha$ ranging from 3.3 to $8.4 \times 10^{-5} \text{ K}^{-1}$ that corresponds to volumetric thermal expansion coefficient ($\alpha_v$) of cubic
perovskites ranging from 9.9 to $25.2 \times 10^{-5} \text{ K}^{-1}$, larger than Cu(In,Ga)Se$_2$ ($\alpha_v = 2.7 \times 10^{-5} \text{ K}^{-1}$) [53] and CdTe ($\alpha_v = 1.4 \times 10^{-5} \text{ K}^{-1}$) [54]. The widely used ITO-coated glass and metal oxide charge transport layers have much lower $\alpha$ values in the range of 0.37 to $1 \times 10^{-5} \text{ K}^{-1}$ [55,56]. Therefore, there is a large thermal expansion difference ($\Delta \alpha$) between the perovskite and substrate.

To achieve high-efficiency PSCs, the perovskite films are typically required annealing at high temperature above 100°C to enhance the crystallinity and reduce defects. Figure 3b shows the processing temperatures of different kinds of perovskites including hybrid organic-inorganic perovskites and all-inorganic perovskites [57]. It should be noted that all-inorganic perovskites even need higher temperatures to form the black cubic perovskite phase compared with hybrid organic-inorganic perovskites. For example, CsPbI$_3$ requires annealing temperatures in the range of 180 to 330°C. There would be a large temperature gradient ($\Delta T$) when perovskite films cool from the annealing temperature to room temperature [31].

The large $\Delta \alpha$ and $\Delta T$ thus lead to the thermally induced strain. That can be quantified using the following equation:

$$
\sigma_{\Delta T} = \frac{E_p}{1-\nu_p} (\alpha_s - \alpha_p) \Delta T ,
$$

where $E_p$ is the modulus of perovskite, $\nu_p$ is the Poisson’s ratio in perovskite, $\alpha_p$ and $\alpha_s$ are the thermal expansion coefficients of perovskite and substrate, respectively. In particular, when a perovskite film forming at high temperature cools to room temperature, it would contract owing to the positive thermal expansion coefficient (Fig. 3c). If a perovskite film is deposited on a substrate with lower $\alpha$, the contact formed between perovskite and substrate during the high temperature annealing process constrains the perovskite from contracting when it cools back to room temperature, leading to tensile strain along the in-plane direction (Fig. 3d). A compressive strain is simultaneously formed in the out-of-plane direction in view of the positive Poisson’s ratio in perovskites.

Another origin of external condition-induced strain is the lattice mismatch between the perovskite and epitaxial substrate. Xu et al. [58] reported the strained epitaxial growth of $\alpha$-FAPbI$_3$ single-crystal thin films on lattice-mismatched halide perovskite substrates. They first tuned the substrate composition of MAPbCl$_x$Br$_{3-x}$, providing a series of host substrates with different lattice parameters. $\alpha$-FAPbI$_3$ was then epitaxially grown on such substrates by inverse temperature growth method. Figure S1b shows the optical images of the as-grown epitaxial $\alpha$-FAPbI$_3$ film. From the high-resolution X-ray diffraction (XRD) patterns of these epilayers (Fig. 3e), the substrate MAPbCl$_x$Br$_{3-x}$ peaks shift to higher diffraction angles as $x$ increases, while the $\alpha$-FAPbI$_3$ peak shift to lower diffraction angles [59,60]. Such a shift to lower angles indicates the increase of the out-of-plane lattice parameter, demonstrating the decrease of the in-plane lattice parameter and then the increase of in-plane compressive strain. When $x$ is above 1.5, the corresponding strain level of the $\alpha$-FAPbI$_3$ film is calculated to be as high as -2.4%. The reciprocal space mapping of strain-free and strained $\alpha$-FAPbI$_3$ films with different lattice mismatch with the substrate (Fig. 3f) further indicates the formation of strain via the lattice mismatch between the perovskite and epitaxial substrate [61].
CHARACTERIZATION OF STRAIN IN PEROVSKITES

Since strain directly alerts the lattice parameters of perovskites, X-ray diffraction (XRD) is thereby a convenient yet effective technique to measure the strain in perovskites [34]. The strain can be calculated by from the shift of XRD peaks:

$$\varepsilon = \frac{d_{\text{strained (hkl)}} - d_{\text{non-strained (hkl)}}}{d_{\text{non-strained (hkl)}}}, \quad (2)$$

The non-strained crystal plane spacing can be obtained from the freestanding perovskite powders prepared by scraping the as-prepared perovskite films from substrates. According to the positive Poisson’s ratio in perovskites, if there is a tensile strain in perovskite film along the in-plane direction, the spacing of crystal planes along this direction becomes smaller; the direction perpendicular to the substrate would be under compressive strain simultaneously, leading to the larger plane spacing along this direction, and vice versa.

There are two different XRD modes: in-plane and out-of-plane measurements. The former measures the plane spacing is of the planes parallel to substrate, while the latter characterizes the plane spacing perpendicular to the substrate, as illustrated in Fig. 4a and b. Micro XRD can further characterize the strain variations on a range of length scales, which can investigate microstructural phase in very small analysis regions about 0.11 mm dimensions [62]. Huang et al. [34] compared the in-plane and out-of-plane XRD of annealed MAPbI$_3$ film (AF) and out-of-plane XRD of scraped MAPbI$_3$ powder (SCP) from annealed film (Fig. 4c). The out-of-plane XRD peak of AF shifted to higher diffraction angle compared to that of strain-free SCP. Such shift demonstrated the smaller plane spacing in the direction perpendicular to the substrate, indicating the compressive strain along this direction. As expected, the in-plane XRD peak of AF shifted to lower diffraction angle compared with out-of-plane XRD peak of AF, determining the larger plane spacing and thus indicating the tensile strain in the in-plane direction of the film.

Grazing incident XRD (GIXRD) has proven to be a powerful technique for characterizing the structural depth profiling of films. That can further be used to study the depth-dependent strain distribution in perovskite films. Specifically, the information from different depths from the surface to bulk is obtained by changing the incident angle; the larger the incident angle, the deeper X-ray penetrates from the surface. Chen et al. [35] recently performed GIXRD to investigate the evolution of in-plane residual strain over the film thickness in mixed perovskite (FAPbI$_3$)$_{0.85}$(MAPbBr$_3$)$_{0.15}$ films, wherein the classical $\sin^2\varphi$ measurement is united with GIXRD (Fig. 4d and e). They fixed the 2θ and varied the instrument tilt angle $\psi$ to obtain corresponding XRD patterns (Fig. 4f). There exists a systematic shift in peak position to lower 2θ as the penetrated depth increased. The slope of the fitting line from the linear relationship between $\sin^2\varphi$ and 2θ further indicates the magnitude of strain (Fig. 4g). The above results demonstrated the gradient distribution of tensile strain in the perovskite thin film, wherein the tensile strain gradually decreases from the top surface to the bulk of the film. This method is also appliable to the characterization of compressive strain in perovskite films, as shown in Fig. 4h and i.

Considering the long-term X-ray radiation-induced phase segregation, the best practice of strain determination by XRD techniques could be the following two methods: (i) choosing single halide perovskites may be better than mixed halide perovskites, whereas phase segregation induced by other factors such as doping and boundary effect under light irradiation may also influence the accuracy of strain characterization; (ii) combination of
in-plane and out-of-plane XRD measurements. Based on the positive Poisson’s ratio in perovskites, if one direction has tensile strain, the vertical direction would have compressive strain, and vice versa. The shift of in-plane XRD peaks therefore is opposite to that of out-of-plane XRD peaks. If there is just a composition-induced peak shift rather than strain-induced shift, the in-plane and out-of-plane XRD results are the same.

Besides the quantitative XRD measurement, there are also other qualitative characterization methods for the measurement of strain in perovskites. Xu et al. [58] applied Raman spectroscopy to study the structure of α-FAPbI$_3$ at different strains between 0% and −2.4%. The peak at 136 cm$^{-1}$ began to split into two peaks at 140 cm$^{-1}$ (main) and 133 cm$^{-1}$ (shoulder) (Fig. 5a and b). The two peaks shifted to 143 cm$^{-1}$ and 130 cm$^{-1}$ as the strain was further increased to −2.4%. This blueshift of the main peak can be attributed to the compression in-plane Pb-I bond, while the redshift of the shoulder peak was due to the stretching of out-of-plane Pb-I bond. Raman spectroscopy measurement can be therefore used to reveal the strain in perovskite films. Ginger et al. [52] characterized the crystal misorientation-induced strain using EBSD (Fig. 5c). The measured local crystal misorientation confirmed the presence of local strain (Fig. 5d and e). The TEM measurement is also used to observe the strain in perovskite films through the measurement of crystal plane distance as shown in Fig. 2d.

As discussed above, there are four widely used methods to characterize the strain in perovskites, including XRD, Raman spectroscopy, EBSD, and TEM. According to equation (2), XRD can provide quantitative measurement of strain through the measured crystal plane spacing. The high-resolution TEM image with micro-area diffraction pattern can also measure the crystal plane spacing and then quantify the strain, whereas it usually suffers from time-consuming and ex-situ process. Raman spectroscopy is only a qualitative characterization method for strain measurement. However, the shifting/splitting/broadening of Raman peaks under different strain conditions can be used to investigate the origin of strain through the analysis of variation in bond rigidity. EBSD that reveals grain structure and internal misorientation in perovskite films can characterize the strain within grains and grain boundaries.

**IMPACTS OF STRAIN ON PEROVSKITES**

As discussed in Origins of Strain in Perovskites, there are several types of strain in perovskites. Particularly, some strains are inevitably residual in perovskites during the film preparation process, such as thermal expansion and lattice mismatch-induced strains. In this section, we briefly review the impacts of strain including both compressive and tensile strains on the physical properties (electronic band structure, electrical properties, defect properties, and ion migration), and stability of perovskites.

**Electronic band structure**

Strain can lead to the lattice distortion and further change the crystal structure of perovskites. This lattice deformation alters the electronic band structure. The calculated band structures of FAPbI$_3$ under tensile, zero, and compressive strains by Chen et al. [35] demonstrate that bandgaps show increase with the strain changing from compression to tension (Fig. 6a). This also leads to the deeper defect levels of perovskites that may act as
non-radiative recombination centers to lower the device performance with the precondition of defect energy levels being not sensitive to strain. Islam et al. [32] further performed high level ab initio simulation techniques to investigate the compressive strain-induced changes in electronic band structure of mixed-cation FA$_{0.75}$Cs$_{0.25}$PbI$_3$. They found that the electronic band structure responds strongly to the compressive strain, wherein the compressed perovskites exhibit narrower bandgaps. These bandgap changes mainly attribute to modulation of the valence band maximum (VBM) rather than conduction band minimum (CBM). As shown in Fig. 6b, the applied compressive pressure of 3 GPa lifts the VBM of FA$_{0.75}$Cs$_{0.25}$PbI$_3$ upward by about 0.2 eV; this strain pushes the CBM upward by only about 0.08 eV. The feasible stress range for tuning the band structure may be under the compressive stress within 2 GPa, corresponding to the strain of 20%~13% considering the modulus of perovskites (approximately 10~15 GPa) [32]. This provides a controlled regulation of bandgap within 100 meV.

This different variation in band edges under strain can be explained through the charge densities of the VBM and CBM. It is well known that the VBM of perovskite consists mainly of the antibonding overlap between Pb 6s and I 5p orbitals, whereas nonbonding Pb 6p orbitals with a very small contribution from I 5p form the CBM. Under the compressive strain, the Pb-I bond lengths becomes shorter, leading to the tilting of the PbI$_6$ octahedra (Fig. 6c). The shorter Pb-I bonds enhance the antibonding overlap between the Pb 6s and I 5p orbitals, thereby increasing the energy of the valence band edge. Although the tilting of the PbI$_6$ octahedra lowers the Pb-I-Pb angles that reduces the antibonding coupling in the VBM, the shorter Pb-I bonds dominated over the distorted Pb-I-Pb angles, ultimately pushing the VBM upward. As for CBM, the nonbonding localized states of Pb 6p orbitals is less sensitive to the shorter Pb-I under compressive strain; there is only a little increase in the overlap between I 5p and Pb 6p nonbonding orbitals. The band edge of CBM therefore also shifts to higher energy but to a smaller extent compared to the VBM, consistent with the experimental results reported by Xu et al. [58] (Fig. S2a). Overall, the application of compressive strain shifts the VBM of perovskites such as α-FAPbI$_3$ and FA$_{0.75}$Cs$_{0.25}$PbI$_3$ to a higher energy that is benefit for the better alignment between perovskite film and hole-transport layer including the widely-used spiro-OMeTAD and PEDOT:PSS, and decreases the bandgap of perovskites.

The change of electronic band structures under strains then alters the bandgap of perovskites. Xu et al. [58] calculated the evolution of bandgap as a function of strain for three α-FAPbI$_3$ lattices with different FA$^+$ organic cation orientations (Fig. 6d). The bandgaps of α-FAPbI$_3$ show red-shift changes with applied strains from tension to compression. Figure 6e exhibits the absorption spectra of strained α-FAPbI$_3$ thin films. The absorption onset redshifts with the increase of compressive strain. Photoluminescence spectra of α-FAPbI$_3$ thin films under different strains from 0% to -2.4% further demonstrated the changes in bandgap (Fig. S2b). The photoluminescence peak shifts from about 1.532 eV under no strain to about 1.488 eV under the compressive strain of -2.4%, indicating a reduction of about 35 meV in the bandgap and demonstrating the feasible strain range for tuning the bandgap of perovskites from experiment [58,63]. Chen et al. [35] also found the similar change in the bandgap of the typical mixed perovskite (FAPbI$_3$)$_{0.85}$(MAPbBr$_3$)$_{0.15}$ under different strains. The measured ultraviolet (UV)-visible absorption spectra and PL spectra under tensile strain, strain-free, and...
compressive strain conditions demonstrated the reduction of bandgap with the increase of compressive strain (Fig. 6f). These strain-induced bandgap changes are consistent with the above first-principles calculations.

**Electrical properties**

The strain-induced alteration of electronic band structures can further change the carrier dynamics of perovskites, since the effective mass of charge carriers is assessed by the band curvature extracted from first-principles calculations. Xu et al. [58] calculated the electron effective masses ($m^*_e$) and hole effective mass ($m^*_h$) under strain from 3% to -3%. Specifically, $m^*_h$ is determined by the curvature of VBM, while $m^*_e$ is from CBM. As discussed above that the VBM is more sensitive than CBM under strains, $m^*_e$ therefore exhibits only a slight variation under strain between 3% and -3%, whereas compressive strain modulates the E-k dispersion of the valence band and greatly reduce the $m^*_h$ (Fig. 7a). They further provided systematical characterizations of the effects of strain on the carrier dynamics of perovskites from experiment. Figure 7b shows the measured Hall effect carrier mobilities of the α-FAPbI$_3$ films under strain from 0% to -2.4%. The films under -1.2% had the highest hole mobility, whereas further increasing the strain led to a drastic decrease of the hole mobility. This may be attributed to the higher dislocation densities arising from the high strain level. Figure S2c shows the calculated carrier mobility from time-of-flight measurements, which was plotted as a function of the strain applied. It exhibits a similar trend to the results measured from Hall effect. In addition to compressive strain, Figure S2d compares the hole mobilities of perovskite films with/without tensile strain measured by space-charge-limited-current (SCLC) method [35], where tensile strain decreased the hole mobility, consistent with the calculated results under tensile strain as shown in Fig. 7a.

**Defect properties**

Metal halide perovskites show extraordinary optoelectronic properties that largely attributes to their unique electronic structure, where antibonding states (Pb 6s-I 5p coupling) locate at the valence band maximum (VBM). This leads to the defect-tolerant properties of perovskites—shallow intrinsic defects in perovskites. This antibonding feature is in contrast with conventional semiconductors such as GaAs [64] and GaN [65] that possess a bonding VBM with deep defect states in bandgap. However, the defects in perovskite films especially at film surface and grain boundaries still induce trap states that dramatically impair both the efficiency and stability of PSCs [14]. The prevalence of intrinsic point defects including vacancies and interstitials correlates closely with their defect formation energy, which directly reflects the density of point defects in perovskites [66,67]. Several groups therefore calculated the formation energies of defects under different strain conditions using DFT. Our group compared the formation energies of halide vacancy under the strains from tensile to compressive. We found that the tensile strain decreases the formation energy of halide vacancies, whereas compressive strain increases their formation energies compared to strain-free perovskites (Fig. 7c) [57]. The low formation energy of defects under tensile strain results in the increase of non-radiative recombination, decreasing the device performance. Islam et al. [32] further calculated the thermodynamic transition levels for vacancy defects of FAPbI$_3$ and FA$_{0.75}$Cs$_{0.25}$PbI$_3$ under different strains. They revealed that the higher pressures above 2 GPa enables a shift of shallow to deep states of iodide vacancies in these perovskites that may act as non-radiative
recombination centers, lowering the device efficiency. Fortunately, at low pressure below 0.5 GPa, the transition state levels of these vacancies remain largely unchanged that would not significantly influence the lifetime of charge carriers.

The impact of local lattice strain on the defect properties of perovskites was recently reported by Sargent et al. [51]. They compared the calculated formation energies of antisites and vacancies in FAPbI$_3$ and mixed CsMAFA perovskites including Cs$_2$MA$_{12}$FA$_{94}$Pb$_{108}$Br$_{55}$I$_{269}$ (CsMAFA') and Cs$_8$MA$_{12}$FA$_{88}$Pb$_{108}$I$_{269}$ (CsMAFA). It should be noted that the local lattice strain in FAPbI$_3$ has been released by partially replacing FA and I with Cs/MA and Br ions. Although no notable difference was observed in the formation energy of Pb-I antisites, there was a significant different in the formation energies of lead iodide vacancies (Fig. 7d). This energy in strained FAPbI$_3$ is as low as about 0.25 eV, while the value in non-strain CsMAFA is increased more than threefold. Therefore, the local lattice strain reduces the formation energies of the lead iodide vacancies in perovskites.

**Ion migration**

Ion migration has been reported to be one of the main causes of photocurrent hysteresis and instability in PSCs [68]. In contrast to the widely observed light-induced ion migration in perovskite films, the impact of strain on ion migration is just beginning to be explored. Our group recently calculated the relative activation energies ($E_a$) for the vacancy-assisted migration of halide ions in perovskites—considering the lowest formation energy of halide vacancy—under biaxial strains from tensile to compressive (Fig. 7e). The activation energies for halide ion migration are 0.547 eV, 0.667 eV, and 0.794 eV for the films under tensile strain (1.5%), no strain, and compressive strain (-1.5%), respectively [57]. The above results indicate that compressive strain increases the activation energy, thereby decelerating the ion migration in perovskites [57,69].

Huang et al. [34] reported the first direct experiment evidence of activation energy ($E_a$) for ion migration in perovskites, which quantitatively describes the degree of difficulty in ion migration. They deposited MAPbI$_3$ films on flexible substrates, and then bent them to be concave or convex to introduce compressive or tensile strain. The temperature-dependent conductivity of the three types of films in the dark was measured. The activation energy for ion migration can be extracted through the Nernst-Einstein relationship:

$$\sigma T = \sigma_0 e^{\frac{-E_a}{kT}},$$

where $\sigma_0$ and $k$ are the constants, $\sigma$ and T are conductivity and temperature, respectively, and $E_a$ is the activation energy for ion migration. $E_a$ is derived from the slope of the ln($\sigma$T) versus 1/T plot. As shown in Fig. 7f, the activation energies for ion migration in tensile-strain, non-strain, and compressive-strain MAPbI$_3$ films in the dark are 0.29, 0.39, and 0.53 eV, respectively. The values under illumination by white light with an intensity of 25 mW cm$^{-2}$ reduces to 0.046, 0.074, and 0.083 eV, respectively. The above measured activation energies demonstrate that perovskite films with tensile strain have smaller ion migration activation energy both in dark or under illumination, whereas compressive strain increases the $E_a$ for ion migration under same conditions, consistent with our calculated results.
Material stability

Point defects—particularly halide vacancies with the lowest formation energy—in perovskites have been regarded as a major source of instability of perovskites due to the following two reasons: (i) point defects have a high affinity for water and oxygen molecules, offering a facile path for oxygen into the perovskite lattice [51]; (ii) defects assist the migration of halide ions, leading to hysteresis in PSCs and halide segregation in mixed-halide perovskites, and subsequently phase segregation [70-73]. As discussed in Electrical Properties and Defect Properties, the formation energy of defects and activation energy for ion migration correlate closely with the strain in perovskites. The impact of strain in perovskites on their stability is therefore an important topic for the stability issues in perovskites.

Huang et al. [34] explored the connection between strain and resulting photostability of perovskite film through depositing MAPbI$_3$ film on flexible substrate. This flexible substrate can be conveniently used to apply external strains from compressive to tensile on perovskite films by bending it in a concave or convex shape. After illumination under white light with an intensity of about 50 mW cm$^{-2}$, they found that the convex film with larger lattice strain had large yellow areas, indicating the decomposition of MAPbI$_3$ into PbI$_2$ as characterized by out-of-plane XRD; the concave film with the smallest strain remained mostly black without any appearance of the PbI$_2$ peak (Fig. 7g). Therefore, the strain impact on the photostability of perovskite film is that tensile strain accelerates the degradation of perovskites under illumination.

Dauskardt et al. [31] carried out similar experiments to investigate the effect of strain on thermal and moisture stability of perovskites. MAPbI$_3$ films deposited on polyethylene terephthalate (PET) with externally applied stresses ranging from -130 to 130 MPa were exposed to either damp air (25°C, 85% RH) or dry heat (85°C, 25% RH) for 24 h (Fig. 7h). They found that films with tensile stress exhibit moisture and thermal instability as seen from the visible degradation with PbI$_2$ formation, whereas films under compressive stress have an improved moisture and thermal stability. The improved illumination, moisture, and thermal stability of perovskite films under compressive strain is attributed to the higher formation energies of point defects and higher activation energies for ion migration under compressive strain as discussed in the previous section.

STRAIN REGULATION METHODS FOR STABLE PEROVSKITES

As discussed above, strain affects physical properties of perovskites that further influences the stability of perovskites consequently. Therefore, regulating strain in perovskites is a novel and effective method to improve their intrinsic stability. In this section, we review the recent efforts to regulate strain for stable perovskites. According to the different origins of strain, these approaches can be divided into two categories: (i) regulation of local strain; (ii) regulation of external condition-induced strain.
Regulation of local strain

Incorporation of A/B/X-site ions

Compositional tuning of perovskites can be used to not only tune the bandgap by halide substitution on the X site, but also improve the thermal stability by Cs and FA substitution on the A site. Recently, new substitution-induced local strain relaxation phenomena have been observed. Priya et al. [67] revealed that the intrinsic instability mechanism of α-FAPbI₃ is the existence of an anisotropic strained lattice in the (111) plane that drives phase transformation into the δ-phase (Fig. 8a). When FAPbI₃ is alloyed with MABr (denoted as FAPbI₃-MABr), the lattice size is reduced due to the smaller ionic radius of MA⁺ and Br⁻ compared to FA⁺ and I⁻, respectively. This substitution balances the lattice strain, leading to the strain relaxation. The sharpening of (111) peaks in FAPbI₃-MABr indicated the strain relaxation of the lattice (Fig. 8b), thereby stabilizing the α-phase of FAPbI₃. Compared to pure FAPbI₃ solar cells, the resultant FAPbI₃-MABr devices exhibited largely improved stability, wherein no drop in efficiency was observed for the device kept under a RH of about 50% for 1000 h (Fig. S3a). Seok et al. [47] further significantly lowered the lattice strain in FAPbI₃ by introducing larger methylenediammonium (MDA²⁺) and smaller Cs⁺ together. They found that the alloyed FAPbI₃ with 0.03 mol fraction of both MDA and Cs cations effectively reduced the local lattice strain as well as the trap desity in PSCs (Fig. 8c). This led to the fabrication of PSCs with 24.2 and 21.6% certified efficiency for small and large (1cm by 1cm) devices. Moreover, the corresponding cells showed excellent thermal stability and maintained almost 80% of their initial efficiency in storage at 150°C for 20 h (Fig. S3b).

Similarly, Chen et al. [74] reported the stress relaxation in perovskite films via A site incorporation. They found that there exists inhomogeneous residual stress distribution across the film thickness direction, wherein the residual tensile stress in the top region of perovskite film was remarkably higher than those at the other depths of the film. This inhomogeneity was attributed to the gradient distribution of cation MA⁺ rather than Cs⁺, FA⁺, Br⁻ and I⁻. The surface residual stress was effectively released to about 50% by introducing two kinds of A-site cations: octylammonium iodide (OAI) and phenethylammonium (PEAI) into the surface of the FA₀.₈₅MA₀.₁₅Pb(I₀.₈₅Br₀.₁₅)₃ films. The mechanism for stress relaxation with OA treatment can be attributed to the incorporation of this large organic cation. This 2D perovskite component mainly dwell at the surface of films to construct soft structural subunits, providing extra structural flexibility to reduce the residual stress (Fig. 8d). The optimized device not only exhibited an improved efficiency of 21.48% (Fig. S3c), but also demonstrated excellent humidity stability and external stress endurance. The devices without encapsulation retained 95% of the initial efficiency after 1000 h under ambient condition (Fig. S3d).

In addition to the above A/X-site incorporation, Sargent et al. [51] demonstrated the incorporation of judiciously selected B-site dopants into the lattice of mixed perovskite crystals that releases the local lattice strain and then increases the formation energy of defects, resulting in an order of magnitude increased stability in ambient air. Specially, they chose Cd owing to its isovalence of Pb while a smaller ionic radius (Fig. 8e). DFT calculated results indicated that Cd incorporation releases the lattice strain and suppresses the formation of I⁻ vacancy. The resultant unencapsulated Cd-containing CsMAFA PSCs thereby showed greatly improved stability. They retained >95% of their initial PCE after storage in the dark in ambient air for 30 days at a relative humidity.
of 50%, whereas CsFAMA PSCs maintained only 60% of their initial PCE (Fig. S3e). They also exhibited an order of magnitude longer operational MPP lifetime under the same conditions when comparing with state-of-art CsMAFA perovskite solar cells (Fig. S3f). Additionally, the incorporation of Cl- also decreased the lattice strain of CsMAFA perovskites due to the small ionic radius of Cl-, similar to the report by Priya et al. [51]. The Cl-containing CsMAFA films exhibited improved moisture and thermal stability, although the incorporation of additional Cl would increase the bandgap that is not desirable for solar cells.

Eliminating inhomogeneity in mixed halide perovskite films

Regarding the local lattice strain originated from the inhomogeneity in mixed halide perovskites, Chen et al. [35] adjusted the perovskite annealing process and proposed a flipped annealing method for the fabrication of perovskite films to modulate the gradient in-plane strain. Specifically, to eliminate the temperature gradient during perovskite film fabrication that induces the compositional inhomogeneity, they added a flipped annealing process to the conventionally annealed perovskite film. This treatment introduced compressive strain and release the tensile strain. GIXRD results showed that the tensile strain was significantly reduced in the sample through this treatment (Fig. 9a and b). Eventually, the optimized strain-free PSC reached a certified efficiency of 20.7% and averaged efficiency of 19.8%, whereas tensile-strained devices only exhibited the efficiency averaged around 18.7% (Fig. S4a and b).

Lattice expansion-induced local strain relaxation

Light-induced structural dynamics has been demonstrated to play a vital role in the physical properties and stability of perovskites as well as their corresponding optoelectronic device performances. In addition to the widely observed light-induced ion migration and halide segregation, illumination can also lead to the relaxation of local strain in perovskites reported by Mohite et al. [75]. They found that continuous light soaking using a standard 1-sun (100 mW cm$^{-2}$) source results in a large and uniform lattice expansion in perovskite films. They considered that photogenerated electron-hole pairs weaken covalent bonds, lead to either less-distorted Pb-I-Pb bonds or elongation of the Pb-I bonds, and finally cause the lattice expansion, resulting in the relaxation of local lattice strain in perovskites (Fig. 9c). This expansion is especially observed in mixed-cation perovskites that are usually strained due to the distorted nature of the lattice with different size cations. GIWAXS measurements showed that all of the diffraction peaks shifted toward lower values of scattering vector $q$ as the illumination time increased (Fig. S4c), corresponding to an isotropic increase in lattice constant (Fig. 9d), so called lattice expansion. The sharpening of Bragg peaks further indicated the relaxation of local lattice strain. This light illumination treatment then increased the performance of p-i-n solar cells using NiO$_x$ as HTL and a FA$_{0.7}$MA$_{0.25}$Cs$_{0.05}$PbI$_3$ active layer from 15% to 20.5% after 2 h of illumination (Fig. S4d). This improvement was attributed to the lattice expansion and a reduction in strain that lower the energetic barriers at the perovskite-contact interfaces. Notably, the devices only exhibited a slow reduction in efficiency under continuous operation at 1-sun illumination for more than 1500 hours (Fig. S4e). However, whether this is light-induced or heat-induced lattice expansion is still under debate. Dauskardt et al. [76] demonstrated that the mechanism for
lattice expansion is heat-induced thermal expansion rather than light-induced expansion through controlling the temperature of perovskite films under both dark and illumination conditions.

**Regulation of external condition-induced strain**

As discussed in External Condition-Induced Strain, the external conditions that induce the strain in perovskites include two types of mismatches: thermal expansion and lattice mismatch between the perovskite and substrate. In this section, we mainly focus on the thermal expansion mismatch, which is the major origin of the strain in perovskite solar cells. According to the above-described correlation between stress and thermal expansion mismatch (see equation 1), there have been several strain modulation strategies reported recently. These approaches can be divided into three categories: (i) lowering the formation temperature of perovskite films to reduce \( \Delta T \) [31]; (ii) using contacting layers possessing similar \( \alpha \) with perovskites to decrease \( \Delta \alpha \) [34].

**Lowering the processing temperature of perovskite films**

The stress is built up during the temperature changes, wherein higher annealing temperature results in larger stress values. In contrast, if a perovskite film is deposited at low temperature even at room temperature, the stress would be decreased dramatically. Huang et al. [34] prepared MAPbI\(_3\) films at room temperature by drying the as-spun MAPbI\(_3\)-DMSO intermediate phase via evacuating for 3 days rather than annealing at 100\(^\circ\)C. They found that the XRD peak positions of this film (pink line) and the nonstrained crystals (green line) are nearly the same, indicating the strain-free state in the perovskite film formed at room temperature (Fig. S5a). They further revealed that this strain-free MAPbI\(_3\) film was still unstrained after being heated at 100\(^\circ\)C for 4 h, whereas the tensile-strain film was still strained even after being annealed at 100\(^\circ\)C for 20 h, indicating that the strain state in perovskites after film formation is insensitive to post-annealing treatment. This can be attributed to the strong interaction between the perovskite and substrate once the perovskite is formed.

Dauskardt et al. [31] also reported that CsFAMA perovskite films formed without any annealing (25\(^\circ\)C) exhibited a stress of \(-10.8 \pm 15.2\) MPa, whereas films annealed at 60\(^\circ\)C and 100\(^\circ\)C resulted in tensile stresses of 20.7 \pm 6.6\) MPa, and 57.6 \pm 4.9\) MPa, respectively (Fig. 9e). These results indicated that the tensile strain in perovskite films can be reduced by lowering the formation temperature of perovskites. They further fabricated perovskite solar cells with the above CsFAMA films in an p-i-n architecture by employing polytriarylamine (PTAA) as the HTL and C\(_{60}\) as the ETL. Figure S5b showed the efficiency of these CsMAFA devices formed at 25\(^\circ\)C, 60\(^\circ\)C and 100\(^\circ\)C, indicating an improvement in PCE with annealing temperature. Although lowering the annealing temperature has been proven to be an efficient way to diminish the tensile stress in perovskite films, this strategy also lowers the device efficiency, which can be attributed to the lower quality of perovskite films fabricated by low-temperature processing.

**Using contacting layers with high thermal expansion coefficients**

According to the equation 1, lowering the thermal expansion difference between the perovskite and contacting layer (\( \Delta \alpha \)) is another way to reduce the tensile strain in perovskites [57]. PSCs typically consist of a stack of multilayer including a substrate with a transparent conducting oxide electrode layer, followed by the ETL,
perovskite layer, HTL, and another electrode layer. The contacting layers of perovskites include substrate and charge-transport layers that usually possess different thermal expansion coefficients (Fig. 3a). Therefore, thermally-induced stress in perovskites can be regulated by choosing suitable contacting layers with similar thermal expansion coefficient to that of perovskites.

Huang et al. [34] deposited MAPbI$_3$ film on a flexible substrate of polyethylene terephthalate (PET) that possesses a thermal expansion coefficient of $2 \times 10^{-5}$ K$^{-1}$ to $8 \times 10^{-5}$ K$^{-1}$, close to that of MAPbI$_3$. The out-of-plane XRD measurements showed that the XRD peaks of the film on PET shifted to lower angles compared to that on ITO/glass (Fig. S5c). This shift indicated that the tensile strain in the MAPbI$_3$ film on PET substrate is much smaller than that on ITO/glass substrate. Dauskardt et al. [31] compared the stress in MAPbI$_3$ films formed at 100$^\circ$C on Si with a low thermal coefficient of $2.6 \times 10^{-5}$ K$^{-1}$ and polycarbonate with a high thermal coefficient of $2 \times 10^{-5}$ K$^{-1}$. As shown in Fig. 9f, the average stresses are 52 and 12 MPa in the films deposited on Si and polycarbonate substrate, respectively. Therefore, the utility of flexible substrates with high $\alpha$ can significantly reduce the thermally-induced tensile stress in perovskite films.

Our group [57] recently reported a strain-compensation strategy that reduces the tensile strain in perovskite films by introducing an external strain from the HTL. The keys for this strain-compensation strategy are the following three aspects. (i) The top HTL layer should have a higher thermal expansion coefficient compared with the perovskite, offering the possibility of compressive strain (Fig. 9g). The poly[5,5-bis(2-butyloctyl)-(2,2-bithiophene)-4,4′-dicarboxylate-alt-5,5′-2,2′-bithiophene] (PDCBT) with a similar chemical structure to poly(3-hexylthiophene-2,5-diyl) (P3HT) is chosen due to its high thermal expansion coefficient (31.5 $\times$ 10$^{-5}$ K$^{-1}$). (ii) The functional layer should have a strong interaction with the perovskite in order to anchor to the lattice and achieve strain offset. The XPS results showed that strong interaction exists between the perovskite and PDCBT through the formation of Pb-O bonds. (iii) The top interface layer should be coated at high temperatures, inducing a compressive strain when cooling back to room temperature (Fig. 9h). Finally, when coating PDCBT at high temperatures, the XRD peaks of as-fabricated perovskite film shifted to higher diffraction angles compared to that of scraped perovskite powder (Fig. S5d). This indicated that the residual tensile strain in perovskite film is successfully compensated through depositing the HTL atop the perovskite at high temperature to introduce compressive strain [57].

We further fabricated three types of PSCs based on different strain-state perovskite films: tensile strain, strain free, and compressive strain for the processing temperatures of the PDCBT HTL coated at 60$^\circ$C, 90$^\circ$C and 120$^\circ$C, with a planar architecture of ITO/TiO$_2$-Cl/perovskite/PDCBT/MoO$_3$/Au. Despite the comparable performance of PSCs under different strains, the photostability under maximum power point (MPP) conditions and thermal stability of PSCs under continuous heating at 85$^\circ$C varied greatly with strain (Fig. S5e and f). The compressive-strain device maintained 95% and 96% of their initial PCEs after continuous MPP operation for 60 h and 1000 h of heating at 85$^\circ$C, respectively, greatly surpassing the strain-free and tensile-strain devices [57].

Another stress-regulation strategy is the direct isolation of the perovskite and substrate due to their mismatched thermal expansion coefficients, and then the addition of soft buffer layer between them. Meng et al. [77] recently introduced polystyrene (PS) into the PSCs as the buffer layer between the SnO$_2$ and perovskite (Fig. 9i). The GIIXRD results showed that residual stresses exist in both the control film (SnO$_2$/perovskite) and experimental...
film (SnO$_2$/PS/Perovskite) as evidenced by the gradual shift of the peaks at 14.0 degree (Fig. S6a and b); however, the PS treated film exhibits a smaller slope than the control film, indicating the less residual stress in the SnO$_2$/PS/Perovskite (Fig. S6c). This revealed that the addition of PS layer can release the thermally induced residual stress in perovskite due to the soft feature of PS with a low glass transition temperature. The resultant strain-free PS-modified PSCs achieved a high efficiency of 21.89% and retained almost 90% and 97% of their initial PCEs after continuously MPP operation for 72 h and after 5 days of “day cycle” stability test, respectively (Fig. S6d).

SUMMARY AND OUTLOOK

We herein presented an overview of the origin, impact, and regulation of the strain in perovskites that govern the performance and stability of perovskite solar cells. The strain in perovskites mainly originates from the local lattice strain and external condition-induced strain including the thermal expansion and lattice mismatch between the perovskite and substrate. The impacts of strain on the physical properties of perovskites, including bandgap, ion migration, hole mobility, defect density, and stability as well as perovskite-based PV efficiency, are complex. For example, the intrinsic interplay between the defect and strain is ambiguous—whether strain induces the generation of defects, or vice versa. Further investigation is needed to uncover the mechanism between strain and their impacts. In addition, shear strain that should be more obvious in large area panels and flexible devices is still rarely investigated, which may be a significant topic for future research. Figure 10 presents the confirmed relationship between strain and these properties. The key impact of strain on perovskites is that tensile strain accelerates the degradation of perovskites that is harmful to perovskites and associated photovoltaic devices; compressive strain improves the intrinsic stability of perovskites and their corresponding PV devices. The aim of strain regulation in perovskites is therefore to release tensile strain and introduce compressive strain. Strain engineering in perovskite films has recently emerged as a central front in advancing perovskite-based optoelectronic device stability and performance.

Despite several years of extensive research on the strain in perovskites, much room remains to advance further the concepts reviewed herein. Although incorporation of A/B/X-site ions is an effective method to relax local strain and increase material stability, this may change the physical properties of perovskites and decrease the device efficiency. Whether light or heat induce the lattice expansion for strain relaxation is still under debate. Other strain regulation methods such as low-temperature fabrication of perovskite films and utilization of flexible substrate can diminish tensile strain in perovskite films, but lower the device performance. The following strategies may be of great importance for further enhancing the stability and performance of perovskite solar cells through strain engineering: (1) Design of mesoporous substrate for the deposition of perovskite layer such as mesoporous TiO$_2$ and ZrO$_2$, wherein the mesoporous layer can extrinsically shield the strain in perovskite layers due to the disruption of the continuity of perovskite films [78]. (2) Exploring new substrates possessing similar thermal expansion coefficients with perovskites. The currently utilized flexible substrates are only limited to polyethylene terephthalate (PET) and polycarbonate (PC). (3) Based on equation 1, one of the methods to decrease tensile strain is to reduce $\Delta\alpha$. Considering the low thermal expansion coefficients of widely-used...
substrates such as ITO-coated glass, decreasing the thermal expansion coefficient of perovskites may be another way to reduce the detrimental tensile strain in perovskite films. (4) Besides the reported external compressive strain from hole-transport layer, other functional layers can also offer compressive strain for perovskites. For example, encapsulant layers especially the encapsulant polymers should not only block air and moisture, but also provide compressive strain due to their high thermal expansion coefficients and high processing temperature. Many encapsulants usually require processing at temperatures of approximately 150°C to cross-link the material and then adhere well to PSCs. (5) Exploring new synthesis methods to fabricate high-quality perovskite films under low processing temperature. Considering that strain regulation is just emerging, and require more knowledge on origins, impacts and engineering, strain engineering in perovskites will continue to help drive higher performance of perovskite solar cells.

SUPPLEMENTARY DATA
Supplementary data are available at NSR online.

FUNDING
This work was supported by the National Natural Science Foundation of China (21922512 and 21875264) and the Youth Innovation Promotion Association of the Chinese Academy of Sciences (2017050).

Conflict of interest statement. None declared.

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Figure 1. Strain origins and its impacts and regulation strategies discussed in this review.
Figure 2. (a) Crystal structure of perovskite in cubic close packing model. (b) A close-packed layer in FAPbI₃, displaying the structural strain. (c) Schematic illustration of the local strain [51]. Copyright 2018, The Springer Nature. (d) The cross-sectional TEM image of (FAPbI₃)₀.₈₅(MAPbBr₃)₀.₁₅-based solar cell. (e-g) The nano-beam electron diffraction patterns [100] zone axis at point e, f and g in a are shown in e-g [35]. Copyright 2019, The Springer Nature. (h) Correlation between misorientation angle and degrees of strain [52]. Copyright 2019, The Elsevier.
Figure 3. (a) Thermal expansion coefficients of functional layers in PSCs including perovskites, substrates, ETLs, and HTLs. (b) Different annealing temperatures of perovskites [57]. Copyright 2020, The Springer Nature. (c) Schematic illustration of the formation of tensile and compressive strain in perovskite films. (d) The calculated annealing temperature-dependent stress in CsPbI$_2$Br and MAPbI$_3$ films [34]. Copyright 2017, The American Association for the Advancement of Science. (e) High-resolution XRD patterns of the (001) peaks of the epitaxial films on different substrates. (f) Reciprocal space mapping with (104) asymmetric reflection of the α-FAPbI$_3$ films [58]. Copyright 2020, The Springer Nature.
Figure 4. Schematics of (a) out-of-plane and (b) in-plane XRD. (c) In-plane and out-of-plane XRD of AF and out-of-plane SCP [34]. Copyright 2017, The American Association for the Advancement of Science. (d) Schematic of the strain measurement using GIXRD, where N0 is the sample normal direction, Nk is the diffraction vector, and ψ is the instrument tilt angle. (e) Diffraction geometries of depth-dependent strain distribution measurement. GIXRD spectra at different tilt angles at the depth of 50 nm for the (f) tensile-strained film and (g) compressive-strained film. Strain distribution in the depth of 50, 200, 500 nm as a function of sin^2φ for the (h) tensile-strained and (i) compressive-strained film. The error bar represents the standard deviation of 2θ [35]. Copyright 2019, The Springer Nature.
Figure 5. (a) Confocal Raman spectra of the epitaxial α-FAPbI$_3$ layer at different strains. (b) Fitting analysis of the Raman peaks in (a) [58]. Copyright 2020, The Springer Nature. (c) Schematics of EBSD measurement on CH$_3$NH$_3$PbI$_3$ thin films. (d) SEM image and (e) inverse pole fig. (IPF) map generated from EBSD of CH$_3$NH$_3$PbI$_3$ thin film with IPF color key. The right column depicts the changes in local crystal orientation along the black arrow in (d) and (e) [52]. Copyright 2019, The Elsevier.
Figure 6. (a) Calculated strain-dependent band structures under biaxial tensile, zero, and compressive strains from first-principle density functional theory-based approaches [35]. Copyright 2019, The Springer Nature. (b) Modification in energy of CBM and VBM. (c) Electronic charge density of the VBM of FA$_{0.75}$Cs$_{0.25}$PbI$_3$ under pressure of 0 GPa (upper panel) and 2 GPa (lower panel) [32]. Copyright 2019, The American Chemical Society. (d) Evolution of lattice volume and bandgap of three α-FAPbI$_3$ lattices with different FA$^+$ organic cation orientations as a function of strain. (e) Absorption spectra of α-FAPbI$_3$ films under gradually increased compressive strains [58]. Copyright 2020, The Springer Nature. (f) UV-Vis absorption spectra and PL spectra of (FAPbI)$_{0.85}$(MAPbBr)$_{0.15}$ under tensile strain, strain-free, and compressive strain conditions [35]. Copyright 2019, The Springer Nature.
Figure 7. (a) Calculated effective masses of carriers of α-FAPbI$_3$ perovskite under different strains from 3% to -3%. (b) Hole mobilities obtained by Hall effect measurements on α-FAPbI$_3$ perovskite film [58]. Copyright 2020, The Springer Nature. (c) Calculated strain-dependent formation energies of halide vacancies [57]. Copyright 2020, The Springer Nature. (d) Calculated formation energies of antisites and Schottky vacancies in FAPbI$_3$ and CsMAFA perovskites [51]. Copyright 2018, The Springer Nature. (e) Calculated strain-dependent activation energies for the vacancy-assisted migration of halide ions [57]. Copyright 2020, The Springer Nature. (f) The activation energy of ion migration of MAPbI$_3$ films under different strains [34]. Copyright 2017, The American Association for the Advancement of Science. (g) Out-of-plane XRD patterns of the films in (a) [34]. Copyright 2017, The American Association for the Advancement of Science. (h) Photographs of MAPbI$_3$ film on PET substrate with externally applied stresses from -130 to 130 MPa aged at 25°C and 85% RH or 85°C and 25% RH for 24 h [31]. Copyright 2018, Wiley-VCH Verlag.
Figure 8. (a) Schematic illustration of the strain in (111) plane of FAPbI$_3$, and strain relaxation after MABr alloying. (b) XRD patterns of FAPbI$_3$ and FAPbI$_3$-MABr [67]. Copyright 2016, The American Chemical Society. (c) Calculated strain in perovskite with a structure of FTO/mp-TiO$_2$/perovskite [47]. Copyright 2020, The American Association for the Advancement of Science. (d) Schematic illustration of stress relaxation with soft and stiff structural subunits [74]. Copyright 2019, The Wiley-Blackwell. (e) Schematic demonstrating the strain reduction through incorporation of small B/X-site ions. [51]. Copyright 2018, The Springer Nature.
Figure 9. (a) GIXRD spectra at different tilt angles at the depth of 50 nm for the strain-free film. (b) Strain distribution in the depth of 50, 200, 500 nm for the strain-free film [35]. Copyright 2019, The Springer Nature. (c) Schematic demonstrating the structure change before (local distortion) and after illumination (lattice expansion). (d) Illumination time-dependent lattice constant [75]. Copyright 2018, The American Association for the Advancement of Science. (e) Measured and calculated stress in MAPbI$_3$ and CsMAFA films formed at different temperature. (f) Measured stress in MAPbI$_3$ films formed at 100$^\circ$C on Si and polycarbonate; insets show the photographs of MAPbI$_3$ on Si and polycarbonate after 45 h of dry heat aging at 85$^\circ$C and 25% RH [31]. Copyright 2018, Wiley-VCH Verlag. (g) Schematic demonstrating the formation of tensile and compressive strains. (h) The calculated PDCBT spin-coating temperature-dependent stress in perovskites within structures of ITO/TiO$_2$/perovskite/PDCBT [57]. Copyright 2020, The Springer Nature. (i) Schematic showing the mechanism of PS buffer layer to release the stress in perovskites. [77]. Copyright 2019, Wiley-VCH Verlag.
Figure 10. Summary of the impacts of various strains on perovskite and device performance.
Table 1. Summary of characterization methods for strain in perovskites.

| Characterization methods | Instrumental accuracy | Measurement criterion | Measurement condition | Reference |
|--------------------------|-----------------------|-----------------------|-----------------------|-----------|
| XRD                      | Quantitative          | Crystalline plane spacing | In-situ              | [34,35,57] |
| Raman                    | Qualitative           | Shifting/splitting/broadening of Raman peaks | In-situ              | [58]      |
| EBSD                     | Qualitative           | Orientation           | In-situ              | [52]      |
| TEM                      | Quantitative          | Crystalline plane spacing | Ex-situ              | [35]      |