Texture Formation in Polycrystalline Thin Films of All-Inorganic Lead Halide Perovskite

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Controlling grain orientations within polycrystalline all-inorganic halide perovskite solar cells can help increase conversion efficiencies toward their thermodynamic limits; however, the forces governing texture formation are ambiguous. Using synchrotron X-ray diffraction, mesostructure formation within polycrystalline CsPbI$_{2.85}$Br$_{0.15}$ powders as they cool from a high-temperature cubic perovskite ($\alpha$-phase) is reported. Tetragonal distortions ($\beta$-phase) trigger preferential crystallographic alignment within polycrystalline ensembles, a feature that is suggested here to be coordinated across multiple neighboring grains via interfacial forces that select for certain lattice distortions over others. External anisotropy is then imposed on polycrystalline thin films of orthorhombic ($\gamma$-phase) CsPbI$_{3-x}$Br$_x$ perovskite via substrate clamping, revealing two fundamental uniaxial texture formations; i) I-rich films possess orthorhombic-like texture ($<100>$ out-of-plane; $<010>$ and $<001>$ in-plane), while ii) Br-rich films form tetragonal-like texture ($<110>$ out-of-plane; $<1\overline{1}0>$ and $<001>$ in-plane). In contrast to relatively uninfluential factors like the choice of substrate, film thickness, and annealing temperature, Br incorporation modifies the $\gamma$-CsPbI$_{3-x}$Br$_x$ crystal structure by reducing the orthorhombic lattice distortion (making it more tetragonal-like) and governs the formation of the different, energetically favored textures within polycrystalline thin films.

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up solution-processed halide perovskite thin films suffer from spatial heterogeneity in their (opto)electronic properties. Beyond identifying processing parameters which can influence grain orientations and their distributions (e.g., material composition, annealing temperature, and annealing time), the underlying origins of texture formation in halide perovskite thin films are still unclear.

The coordinated alignment of structural domains across multiple halide perovskite crystals requires two central ingredients: i) intrinsic domain formation across multiple grains, either through widespread nucleation or rapid spreading, and ii) introduction of an external anisotropic field which can preferentially select for the formation of certain domains over others. Due to their improved thermal and chemical stability, all-inorganic cesium lead halide perovskites are envisioned to champion long-term, stable devices. However, CsPbI$_3$-based perovskites form a stable yellow nonperovskite structure (δ-phase) at room temperature (RT), requiring high-temperature processing (≥320 °C) to access their black perovskite phases (cubic, α; tetragonal, β; orthorhombic, γ) before being stabilized for applications at RT. Starting from the archetypical high-temperature cubic phase in Figure 1A, octahedral distortions reduce the lattice symmetry and form structural domains upon cooling, being driven by thermal contraction and the development of spontaneous strains within the crystal. High temperature processing of halide perovskite is typically paralleled by large changes in the final thin-film (micro)structure and morphology. For instance, annealing induces grain coarsening (Figure 1B) which is often tailored toward evolving smaller nanocrystals into larger micrometer-scale grains, to reduce the parasitic influence of grain boundaries.

First-order high-temperature phase transitions within halide perovskites unavoidably imply phase intergrowths and coexistence. Within this context, it is their inherent polymorphic nature and relatively soft crystal structure (tendency to form heterojunctions) which will govern complex mesostructure formations, like the thin-film texture illustrated in Figure 1C. Thus, a relatively simple perovskite crystal structure may exhibit immense structural complexity at larger length scales, influencing important features—like micrometer carrier diffusion lengths—at the thin-film level.

Herein, we examine how changes in the crystal symmetry of free-standing polycrystalline all-inorganic halide perovskites drive common grain orientations and anisotropy during annealing and subsequent cooling, manifesting as texture within polycrystalline thin films. Using synchrotron X-ray diffraction (SXRD) we decouple the development of crystallinity (i.e., the enlargement of crystal grains) and domain ordering (i.e., the collective alignment of octahedral tilts within separate grains) in ensembles of CsPbI$_2$Br$_{0.15}$ microcrystalline powders during thermal treatment, which represents two key processes: i) heating randomly oriented as-grown grains causes them to sinter and coarsen before transitioning to the black perovskite phase, where ii) cooling of the α-phase perovskite polycrystalline assembly introduces common distortion directions within the individual crystals, triggered by the β-phase transition. For thermally treated γ-CsPbI$_{3−x}$Br$_x$ thin films (formed by quenching to RT), a thermal contraction mismatch with the underlying transparent substrate (i.e., glass) biaxially strains the material promoting two distinct uniaxial texture formations, depending on the Br content. We examine thin-film texture using synchrotron grazing-incidence wide-angle X-ray scattering (GIWAXS) and connect the different texture expressions to Br-induced suppression of orthorhombic distortions in γ-CsPbI$_{3−x}$Br$_x$ perovskite, a feature which is shown to be dominant over all other processing parameters, i.e., annealing temperature, choice of device substrate, and film thickness.

Needle-like, microcrystalline powders of δ-CsPbI$_2$Br$_{0.15}$ are grown by drop casting (Figure S1, Supporting Information) which preserve bulk-like phase properties during high-temperature restructuring, without exposure to extrinsic influences like hetero-interfaces or surface-driven effects (i.e., the change in surface tension occurring in nanocrystals). Substituting 5% of the I sites with Br expands the temperature range for which the metastable perovskite phase exists and can be studied in situ during phase restructuring.

Mesostructures formed across large grain populations, i.e., with respect to their size, shape, and crystallographic orientation, can arise over lengths of hundreds of nanometers. For SXRD experiments employing large-area 2D detectors, this information is encoded in the shape, width, and azimuthal angular distribution of Debye–Scherrer diffraction rings (Figure 1B,C). SXRD patterns are recorded while heating CsPbI$_2$Br$_{0.15}$ powders up to 597 K and cooling back to RT, yielding several phase transitions (Figure S2, Supporting Information), as displayed in the T-t profile in Figure 2A. The δ-CsPbI$_2$Br$_{0.15}$ powder first transitions...
into a black phase at 568 K, with the cubic $\alpha$-phase maintained until cooling where it undergoes a tetragonal distortion, before again transitioning back to a stable $\delta$-phase.

Figure 2B contains SXRD frames recorded from both the $\delta$-phase and the $\alpha$-phase before any common grain orientations form (full images in Figure S3 in the Supporting Information), yielding near homogeneous Debye–Scherrer rings due to the inherent disorder of grain orientations at this time. Figure 2C displays an enlargement of the frames to better track the evolution during thermal treatment. Local spottness develops within the diffraction rings of the yellow phase during heating, indicating grain coarsening (Figure 1B). Thermally induced changes to the morphology is confirmed by scanning electron microscopy (SEM) images of the CsPbI$_{2.85}$Br$_{0.15}$ powders heated to different temperatures, showing widespread sintering of the needle-like microcrystals (Figure S4, Supporting Information).

From the yellow-to-black phase transition up to the maximum temperature of 597 K (Figure 2C), no further changes related to ring spottness and grain coarsening are observed. Cooling, however, induces large changes in the Bragg azimuthal distributions of the SXRD rings; the scattering image recorded at 550 K exhibits a more single-crystal-like pattern, with the formation of highly inhomogeneous and even incomplete scattering rings (i.e., with some pixels in the path of the scattering rings not exposed) and several intense hotspots. Further, regions of high signal intensities are found to bunch and smear, similar to the signals recovered from heavily twinned multiple single crystals. To rule out extensive grain growth as the origin for this rapid change, electron backscatter diffraction (EBSD; Figure S5, Supporting Information) measurements confirm that each individual crystal within the partially fused bundles keeps its original microstructure and orientation (i.e., no coherence between grains). This suggests grain growth (Figure 1B) is not the dominant cause of changes within the X-ray scattering ring distribution. Given the relatively large number of individual micrograins contained in the scattering volume (we estimate as many as ten to twenty thousand grains), such organization is intriguing. We suggest the likely cause is related to the coalignment of structurally coherent domain blocks at several length scales; from sub-micrometer blocks within grains, to large grains forming the polycrystalline aggregates, i.e., micrometers and tens of micrometers.

An azimuthal profile analysis is used to evaluate the distribution of pixel intensities across a portion of the (200) black phase scattering ring during cooling (Figure S6, Supporting Information). Cooling from 583 K causes the pixel intensity distribution to shift closer to the detector baseline, as the Debye–Scherrer rings become more intermittent. The degree of inhomogeneity

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**Figure 2.** a) In situ SXRD T–t profile ($\lambda = 0.95774$ Å) of CsPbI$_{2.85}$Br$_{0.15}$ through a high-temperature yellow-to-black phase transition (565 K) and cooling ramp. b) Representative large-area SXRD images recorded from the yellow phase and high-temperature black phase. c) Enlarged portions of the scattering patterns followed at different stages of the heating (red arrow) and cooling (blue arrow) ramp, corresponding to the green area identified in (B). Note that the images series are separated into yellow (top) and black phase patterns (bottom) at different stages of the same thermal cycle shown in (A).
in the intensity distribution across (200) is directly influenced by the size, number and orientation of grains, and is tracked by performing polar integrations over a fragment of the scattering ring (Figure S6, Supporting Information). The results in Figure 3A show no development in the azimuthal distribution of the diffracted intensity while heating the black phase (i.e., negligible grain growth), however upon cooling there is a rapid increase in pixel shading (i.e., the fraction of azimuthal bins for which the GIWAXS intensities do not exceed the background). The sudden absence of X-ray signals incident on portions of the (200) Bragg diffraction ring indicates the loss of a good powder average, as one would expect for a randomly oriented polycrystalline material.

An analysis of the normalized lattice spacing evolution reveals that domain ordering correlates with the tetragonal distortion formed in the cooling perovskite unit cell. With $\beta$-phase formation appearing to be the trigger, grain orientation will be mediated by intergranular stimuli within the heavily interfaced polycrystalline network (Figure S4, Supporting Information). We suggest that intergranular tension (the strain exerted by noncoherent interfaces with different thermally expansions$^{31}$), or the highly dynamic nature of the halide perovskite crystals during restructuring$^{35}$ as potential mechanisms for domain selection; however, the preferred directions for grain orientation are not fixed without an external anisotropic field.

We next consider how preferential directions in grain orientations might form under different scenarios. A tetragonal distortion of an isotropic cubic unit cell involves domain formation in one of three equally likely directions (Figure 4A), however the SXRD patterns recorded from free-standing perovskite polycrystalline powders reveal an inherent tendency toward the formation of organized grain orientations during cooling of fused polycrystals (Figure 4B). In the presence of a permeating anisotropic field (i.e., via a strained planar interface), Figure 4C schematically depicts how anisotropic grain orientations are expressed at scale, to form a statistically distributed polycrystalline texture throughout an entire thin-film grain population.$^{25}$ This we study in detail below.

With crystal symmetry revealed to drive texture formation, Figure 4D assesses the changes taking place in the normalized lattice of $\gamma$-CsPbI$_{1-x}$Br$_x$ perovskite with increasing Br content. A systematic reduction of the crystal volume is imposed by the replacement of I sites with relatively smaller Br anions.$^{36}$ The relative changes in the normalized lattice parameters are linked to the redistribution of spontaneous strain measured as deformations to the parent high-symmetry unit cell, induced by the phase transitions.$^{37}$ For example, with increasing Br content, the difference between the $a$- and $b$-axes (representing the spontaneous orthorhombic distortion) is significantly reduced. This strongly mirrors the effect of heating a metastable RT $\gamma$-CsPbI$_3$ perovskite toward higher temperatures,$^{15}$ which increases both the crystal symmetry (becoming more tetragonal-like and then cubic) and the thermodynamic stability.

For phase transitions in which the high-symmetry $\alpha$-phase is reduced to a degenerate $\gamma$-phase, the number of distortion components can be expressed in terms of symmetry-adapted strains.$^{38}$ The degenerate symmetry-breaking distortions are thus divided into the tetragonal ($e_{\text{tet}}$) and orthorhombic ($e_{\text{orth}}$) strains, manifesting the $\beta$-phase and $\gamma$-phase, respectively. These quantities are calculated relative to an undistorted cubic unit cell, $a_0$, which is estimated by taking the cube root of the normalized unit cell volume. It follows that the spontaneous strain components are defined as: $e_1 = (a - a_0)/a_0$, $e_2 = (b - a_0)/a_0$ and $e_3 = (c - a_0)/a_0$, where $a$, $b$ and $c$ are the normalized lattice parameters of the CsPbI$_{1-x}$Br$_x$ orthorhombic phase. The separate strain components contributing to the lattice distortions are then given by: $e_{\text{tet}} = e_1 - e_2$ and $e_{\text{orth}} = (2e_1 - e_1 - e_3)/\sqrt{3}$. A factor of $\sqrt{3}$ is included here to ensure that the two strains are on the same scale. Figure 4E,F presents the two spontaneous strain components and reveal that $e_{\text{orth}}$ is significantly reduced in $\gamma$-CsPbI$_{1-x}$Br$_x$ with rising Br incorporation ($\approx$80% for $x \geq 2$), while $e_{\text{tet}}$ remains unchanged.

Next we explore texture formation within high-temperature processed polycrystalline $\gamma$-CsPbI$_{1-x}$Br$_x$ thin films (Figure S1, Supporting Information) using synchrotron-based GIWAXS. Starting from a randomly-oriented, as-grown $\alpha$-CsPbI$_3$ thin film (Figure S8, Supporting Information), thermal heating and quenching generates a metastable black phase and introduces biaxial strain$^{25}$ due to the large mismatch in thermal expansion of the perovskite layer ($\approx 50 \times 10^{-6} \, \text{K}^{-1}$) and the underlying glass substrate ($\approx 4 \times 10^{-6} \, \text{K}^{-1}$). Figure 5A presents a 2D GIWAXS intensity image of a quenched RT $\gamma$-CsPbI$_3$ thin film rendered to correspond with the scattering azimuthal angle (off-center detected image with wider in-plane scattering range.
is shown in Figure S9 in the Supporting Information). Selected Bragg peak intensity distributions are provided in Figure S5B, showing (002) and (020) reflections reach their maxima in-plane, with (200) pointing normal to the thin-film surface. The uniaxial texture confines the (110) peak maximum to point out-of-plane in a bimodal fashion (two maxima separated by 90°). The azimuthal distribution of (200)—which exhibits no clear peak—is well-described using two adjacent cumulative distribution functions (CDF), to create a step-like probability function centered at $\chi = 0°$: $\phi_{(002)}(\chi) = A \times \left[1 - \left(CDF_a + CDF_b\right)\right]$, where the normalization constant $A$ and the error function, $\text{erf}$, describe the mean deviation from the substrate normal, i.e., $2\mu = \text{full-width at half-maximum (FWHM)}$, and $\sigma$ is the variance at edges, governing the skew rate. Fitting the GIWAXS data recorded from our $\gamma$-CsPbI$_3$ thin films with this distribution function, $\phi_{(002)}(\chi)$, yields $\mu = 29°$ and $\sigma = 5.1°$. We hereon identify the texture found in thin-film $\gamma$-CsPbI$_3$ as orthorhombic-like, $\phi_{\text{orth}}$; see Figure 5C.

To investigate whether the resultant $\gamma$-CsPbI$_3$ texture is a general one, similar analyses were performed on thin films with different thicknesses (Figure S10, Supporting Information) and deposited on different inorganic device-ready substrates (Figures S11 and S12, Supporting Information). Disregarding relatively rough mesoporous titanium oxide (m-TiO$_2$) substrates (examined below), both the substrate choice (all with thermal expansion coefficients $c_{\text{orb}}$ [32,39,40] much lower than the perovskite layer; ranging $−4 \times 10^{-6}$ to $10 \times 10^{-6}$ K$−1$) and thin-film thickness (from 140–900 nm) have little influence on the final $\gamma$-CsPbI$_3$ thin-film texture. This suggests that the texture formation is effectively translated from the strained interface through the whole thin-film layer, even when it is comprised of multiple stacked grains, as is the case with our thicker samples. Together, these additional experiments support a crystal symmetry-driven mechanism for the texture formation.

Br incorporation in $\gamma$-CsPbI$_{3−x}$Br$_x$ leads to a substantial reduction in the $\epsilon_{\text{orb}}$ strain component and a subsistence of $\epsilon_{\text{tet}}$ (Figure 4E,F). Thus, one can expect that Br-introduction can modify the polycrystalline texture formed in Br-rich CsPbI$_{3−x}$Br$_x$ thin films. Figure 5D presents a 2D GIWAXS intensity image of a thermally quenched $\gamma$-CsPbI$_{Br_x}$ thin film with the Bragg intensities in the azimuthal domain. Off-center images, showing the (004) peak, are shown in Figure S13 in the Supporting Information. The texture exhibited in thin-film $\gamma$-CsPbI$_{Br_x}$ is different to the pure triiodide system, having <110> normal to the planar surface in a relatively narrow distribution (FWHM of Lorentzian is $≈10°$). Consequently, the uniaxial texture limits the <110> and <001> directions to be in-plane. This results in the (200) and (020) lattice planes—which are approximately equal—to appear in a
bi-modal fashion at an azimuthal angle of ±45°. Likewise, the (112) peak in a tetragonal-like system arises close to the same 2θ and χ scattering angles, which is why it remains coupled in the corresponding intensity plot in Figure 5E. This texture formation is identified as tetragonal-like, ϕ_{tet}, and is displayed in Figure 5F. Tetragonal-like texture has been widely reported within hybrid organic-inorganic lead halide perovskite thin films[14,20,41] which tend to adopt tetragonal or tetragonal-like perovskite crystal structures. Moreover, a ϕ_{orth} texture was recently reported in tetragonal β-CsPbI_{3} thin films, indicating that the crystal symmetry, rather than the composition, is most influential.

In line with its crystal symmetry-driven origins, the direction and distribution of ϕ_{orth} is found to remain steady across a broad range of annealing temperatures (Figure S14, Supporting Information). Conversely, an analysis of γ-CsPbI_{3}Br thin films with an intermediate Br content reveals a hybrid texture formation (Figure S15 in the Supporting Information: simultaneous contributions of both ϕ_{orth} and ϕ_{tet}). However, this dual texture is strongly dependent on the annealing temperature: higher annealing temperatures promote a relatively stronger ϕ_{tet} expression and narrow its angular distribution (Figure S16, Supporting Information). Similar dual textures have been reported previously in CsPbI_{1.8}Br_{1.2} solar cells.[42]

The bimodal nature of the texture formation in quenched, black CsPbI_{3−x}Br_{x} thin films, and its connection to crystal symmetry, suggest that these directions represent fundamental, energetically favored formations. For the case of γ-CsPbI_{3} thin-films, anisotropic thermal expansions[13] arises because the tilting distortions are different. Here the normalized b- and c-axes of the γ-phase CsPbI_{3} experience the smallest lattice reduction.
at RT, while the a-axis contracts much faster and ends up the smallest. We suggest that the introduction of tensile in-plane strain after quenching favors distortions that approximately align the b- and c-axes in-plane—directing the a-axis out-of-plane—to minimize the tensile strain introduced after cooling.

Next, we elucidate how strain-driven energy differences might arise in thermally treated CsPbI$_3$-Br$_x$ thin films using periodic density functional theory (DFT; see the Experimental Section and Figure S17 in the Supporting Information) to calculate the ground state energy difference of α, β, and γ phase CsPbI$_3$ perovskite. Applying 1% strain along different crystal planes (Figure S18 and Table S1, Supporting Information) shows that strains the (001) plane in both the β and γ phases is energetically unfavored, and the c-axis should therefore preferentially align parallel to the interface, in agreement with experiment. Further, the length of the free lattice vector is always smaller than the strained lattice vectors (Table S1, Supporting Information), with the shortest lattice vector oriented perpendicular to the strain. Additionally, biaxially straining the (100) plane will be preferred over the (111) plane for the β-phase and the (112) and (010) planes for the γ-phase. This supports the notion that the longer b- and c-axes of the primitive β and γ unit cells will orient parallel to the interface. While this agrees directly with the experimental orthorhombic-like texture for γ-CsPbI$_3$ (Figure 5C), it is unexpected for the tetragonal-like texture of γ-CsPbBr$_2$ (Figure 5F).

To understand this difference, it is important to investigate the ground state energy difference between the (100) and the (110) planes for both β and γ-CsPbI$_3$, taking into account that only the pure iodide material was theoretically investigated (Figure S18, Supporting Information). Starting in the γ-phase, Figure S18 in the Supporting Information demonstrates that biaxially straining the (110) plane in CsPbI$_3$ is about 19 meV per formula unit (p.f.u.) less favorable than biaxially straining the (100) plane. By decreasing the magnitude of the orthorhombic strain $e_{\text{orth}}$, this difference in ground state energy decreases to about 11 meV p.f.u. for the β-phase of CsPbI$_3$. This observation can be explained given that the a-axis is significantly shorter than the b-axis for the γ-phase while they are equal for the β-phase. The less pronounced preference for the (100) plane upon converting from the γ to the β-phase in CsPbI$_3$ can therefore be correlated with the reduction in magnitude of $e_{\text{orth}}$. While biaxially straining (110) never becomes more favorable than straining the (100) plane for CsPbI$_3$—even without orthorhombic strain an 11 meV p.f.u. difference remains—this clearly shows that reducing the orthorhombic strain is crucial to increase the likelihood of observing the tetragonal texture. Given that Figure 4E experimentally demonstrates that the magnitude of the orthorhombic strain decreases with increasing Br incorporation, this observation forms a possible explanation of why a tetragonal-like structure is obtained in tetragonal-like γ-CsPbBr$_2$. Further, this hints that the total energy will depend on a thermodynamic surface term, as a high-symmetry plane like (110) is a preferred interface plane while in the perovskite phase. Within this scenario, it will be a trade-off between the preferential alignment due to biaxial strain and the interfacing surface that determines the observed texture.

In champion halide perovskite solar cells, the use of mesoporous titanium dioxide (m-TiO$_2$) substrates is commonplace, which has, by design, a relatively rough surface morphology (Figure S11, Supporting Information). We evaluated the texture formation in device-ready FTO/c-TiO$_2$/m-TiO$_2$/CsPbI$_3$-Br$_x$ thin films over a solar-friendly compositional range; 0 ≤ x ≤ 1.2. Figure S19 in the Supporting Information focuses on the (110) peak evolution with rising Br, showing that the rough surface of m-TiO$_2$ introduces a disorderly, hybrid texture distribution of both φ$_{\text{rot}}$ and φ$_{\text{ext}}$ in CsPbI$_3$-Br$_x$ thin films, which is quickly shifted toward φ$_{\text{rot}}$ by increasing the Br content. This suggests that the added surface roughness does not allow the interfacial forces at the junction to be uniform enough to clearly discriminate between different texture formations.

In summary, we have systematically studied texture formation within polycrystalline thin films of all-inorganic metal halide perovskite CsPbI$_3$-Br$_x$ using synchrotron X-ray diffraction, structure-sensitive electron microscopy and DFT. First, through the examination of free-standing polycrystalline ensembles, shared nanograin orientations are shown to be an inherent feature within soft, polymorphic perovskites, being triggered by thermally induced distortions within the unit cell. When expressed at scale within typical solution-processed thin-film device architectures (i.e., atop transparent glass substrates), we discovered two distinct texture formations manifesting within CsPbI$_3$-Br$_x$ thin films. I-rich films are shown to lead to orthorhombic-like texture (<100> out-of-plane; <010> and <001> in-plane), while Br-rich films form tetragonal-like texture (<110> out-of-plane; <110> and <001> in-plane). Following an exhaustive exploration of material processing parameters like substrate chemistry and roughness, film thickness and annealing temperature, the amount of Br incorporation in γ-CsPbI$_3$-Br$_x$ thin films is revealed to be the central governing factor toward producing different texture formations, suggesting they are energetically favored formations. This is because Br incorporation modifies the γ-CsPbI$_3$-Br$_x$ crystal structure by reducing the orthorhombic lattice distortion (making it more tetragonal-like), which changes the domain selection criterion coordinating texture formation at the thin-film level. Our findings help to unlock the interpretation of several anomalous reports of anisotropic thin-film behavior in polycrystalline perovskite thin films, typically derived using linear or single-pixel X-ray scattering detection. Comparing our findings with wider research on hybrid organic-inorganic halide perovskites, like the popular hybrid methylammonium and formamidinium lead halide perovskites, the insights revealed here unify texture formation within thermally processed perovskite thin films. Being able to fine tune specific, energetically favored polycrystalline texture formations will ultimately lead to engineering device-ready materials. Large-area SXRD appears best suited to decipher the statistical information encoded in the fine nanoscale grains and polycrystalline thin-film texture. Only with precise statistical data, like that depicted in Figure 5, can the influence of texture on carrier diffusion and transport within thin films be accurately quantified or understood, leading to improved device performance.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.
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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

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