able to reach a break-even point (net present value, NPV > 0) in 4–6 years in spite of a rather high battery system cost (€380/MWh) assumed. For comparison, the best single-use application (spot market trading) would reach a positive NPV not until the 9th year of operation. The operating profit from the multi-use is slightly lower than the sum of the single-uses due to trade-offs in different power and energy capacity sharing.

As their analysis was limited to four revenue streams only (peak-shaving, self-consumption, frequency reserve, spot market trading), excluding, e.g., supply capacity, power distribution, transmission, customer energy management applications, and other ancillary services, the above estimates may be conservative, though not all of these may be captured by the present Li-ion technology.[3] On the other hand, decreasing returns could be expected with a larger number of similar systems instead of one agent like in the present study.[8] The practical control of a battery for allocating dynamically its capacity to multiple uses may be challenging, though the authors show that a quite long switching time up to half of

day could be allowed for reallocating without severely affecting the profitability. Whereas for a sequential allocation strategy, i.e., providing exclusive service of one application at a time, the profits start to drop when the switching time exceeds a few hours.

Though dynamic stacking of multiple applications could technically be feasible, the present regulatory framework prohibiting vertical bundling in many countries may inhibit using a battery storage cross the transmission and distribution system. This may require updating present electricity market legislation to allow distributed storage systems to be located in the intersection of the transmission and distribution system and serve the needs of both parts of the grid system. Shared storage between the Transmission and Distribution System Operator could be a solution to the allocation problem. Succeeding in releasing the full potential of battery storage would definitely help in decarbonizing the energy system.

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Visualizing the Invisible in Perovskites

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Halide perovskites are promising for a spectrum of energy applications including photovoltaics. Decisively characterizing microstructures of perovskites is critical yet challenging due to their soft characteristics. Recently, Rothmann et al. employed low-dose, low-angle annular dark-field scanning transmission electron microscopy to image perovskite thin films at the atomic scale, unveiling key structural details that are invisible to previous observations. This study opens the door to revealing microscopic structure-property-performance relationships in perovskites and beyond, guiding the reinvention and optimization of energy devices.

The increasing energy consumption of modern society drives an urgent demand for clean and renewable energy generation beyond fossil fuels. Among a spectrum of alternative energy sources including wind, hydro, solar, and nuclear powers, photovoltaics (PVs) have been considered as one top technology candidate because they could potentially convert solar energy of

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about ten thousand exajoules annually into electricity that will be conveniently distributed for the everyday activities of human beings. Therefore, since the 1990s, there has been a continuous, vast effort to develop high-efficiency and low-cost PVs in numerous countries such as the United States, China, Australia, and so on. While silicon (Si) has been proven to be able to serve in PV panels efficiently, and they are robust in real-world applications, the energy payback time of Si PVs can be more than two years, and their production involves high-temperature synthesis and a hazardous purification process, inevitably imposing adverse effects on the environment. In this regard, the field of solar energy is intensively searching for next-generation PV materials that not only exhibit favorable optoelectronic properties for high device performance but also are easy and cheap for industrial fabrication. One most significant steps that meets this demand is the intriguing discovery of halide perovskites (HP)-based PV materials, drawing phenomenal attention across the academic, industrial, and governmental sectors.

The first perovskite solar cell (PSC) was reported by Miyasaka and coworkers with an efficiency of only 3.8%,[1] but the following decade witnessed unprecedented growth and expansion of this emerging research area, leading to a swift evolution in device efficiency to 25.5%.[2] This progress is largely driven by the innovation and optimization of device fabrication processes.[3] Nevertheless, it now appears that the efficiency progress of PSCs has become relatively saturated, and the long-term stability is raised as a significant concern. To address these critical issues, in parallel to the device development, there is an ongoing effort to resolve the fundamental structure-property-performance relationship at multiple scales in HPs, the results from which are expected to guide the materials and device engineering to improve

PSCs.[3] At the center of this research effort is a decisive, accurate characterization of microstructures and defects in HP thin films because numerous studies have shown that microstructures and defects play a significant role in influencing both carrier dynamics and mass transport in HP thin films, determining efficiency and stability of PSCs, respectively.[4] In this regard, it becomes an urgent need to develop reliable, powerful characterization techniques. While common tools like optical microscopy, scanning electron microscopy (SEM) and atomic force microscopy (AFM) have been commonly used for morphological determination of HPs, they are generally limited to probing the sample surface without revealing details of materials' interiors. It is well known that transmission electron microscopy (TEM) is capable of imaging at a much higher resolution, and it is well established for characterizing conventional materials. However, as a new class of "soft" semiconductors, HPs are prone to degradation upon exposure to electron irradiation, presenting an obstacle for high-quality TEM imaging.[5] While there is a handful of promising studies that involve high-resolution TEM of freestanding HP nanomaterials based on specific methods such as cryogenic TEM,[6] atomic-scale structures of HP thin films are rarely revealed, and thus the true mechanisms responsible for the PSC performance and stability are still much obscured.

Now, remarkable progress has been made by Rothmann et al.[7] and was published in a recent issue of *Science*, demonstrating the imaging of microstructures of HP thin films with atomic-level spatial resolution ($\sim 10^{-10}$ m) using aberration-corrected scanning TEM (AC-STEM). STEM is a typical mode of TEM working with a focused electron probe instead of the parallel electron beam used in conventional TEM. With the recent development of aberration correctors, the electron probe is

focused down to the atomic or even sub-atomic scale to observe structural details. The pixel scanning probe mechanism makes STEM able to directly reflect the accurate real-space position of atoms. Furthermore, STEM annular dark field (ADF) imaging has a contrast sensitivity to the atomic number (Z), enabling us to distinguish different types of atoms in a facile way. Although STEM is widely used for characterizing functional materials including oxide perovskites, it has been rarely applied to HPs. Rothmann et al.[7] chose a thin film sample of formamidinium lead iodide ($FAPbI_3$) perovskite, the most typical HP in state-of-the-art PSCs.[8] They employed the low-dose, low-angle annular dark field (LAADF) STEM imaging, a technique that not only maintains the sensitivity to Z variations but also collects a higher fraction of the scattered electrons for low-dose imaging than conventional high-angle annular dark field (HAADF) STEM. With this technical development, several intriguing materials-science findings on HP microstructures were demonstrated.

Rothmann et al.[7] show the direct atomic-scale imaging of various crystal defects in the $FAPbI_3$ HP thin film with zero-dimensional (0D) to three-dimensional (3D) structural dimensions (see Figure 1). For 3D defects, they observe that PbI_2 nanodomains within in the perovskite phase. Surprisingly, a coherent transition boundary with low lattice misfit and strain is found between the remnant PbI_2 (a precursor phase) and $FAPbI_3$ lattice. This may suggest that the epitaxial-like PbI_2 nanodomain could serve as a seed during the HP solution growth, and such structural coherence could explain why a small amount of excess PbI_2 doesn't negatively affect the PSC performance. For two-dimensional (2D) defects, grain boundaries and stacking faults are prominently observed. Rothmann et al.[7] show a well-defined crystallographically continuous grain boundary structure with a sharp interface and
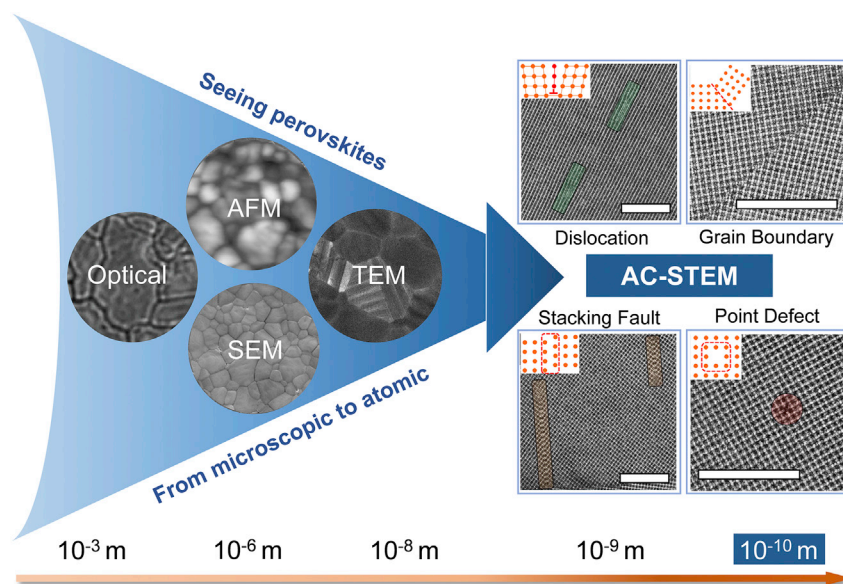
Figure 1. Schematic Illustration Showing the Resolution of Structures of Halide Perovskite Semiconductors from Microscopic to Nanoscopic to Atomic Scales Based on the Progression of Characterization Techniques

The optical microscopic image is adapted from Li et al., 2018[9] with permission from the American Chemical Society. The AFM and SEM images are adapted from Song et al., 2020[10], and the TEM image is adapted from Rothmann et al., 2017,[11] both under a Creative Commons Attribution v4.0 International license. The AC-STEM images are adapted from Rothmann et al., 2020[7] with permission from the American Association for the Advancement of Science (AAAS). The scale bars shown in the AC-STEM images are all 10 nm.

minimal lattice distortion. The near-120° triple boundaries represent the majority of observed grain boundaries in FAPbI$_3$. For the stacking faults, they exhibit a shift of a half unit cell and connect Pb-I columns with I$^-$ columns. Also, one-dimensional (1D) defects of edge dislocations are revealed, and they are dissociated in a direction perpendicular to their glide planes. Furthermore, Rothmann et al.[7] show a preliminary identification on the atomic structure of aligned 0D defects of vacancies on the Pb-I sublattice. They also point out that an intermediate structure forms with the partial loss of FA$^+$ ions before the FAPbI$_3$ completely deteriorates to PbI$_2$, and this implies that an ordered FA$^+$-deficient intermediate structure may form as a result of facile ion migration.

With such comprehensive imaging of defect structures, mechanistic investi-gations of optoelectronic properties and chemical stability of HPs will be facilitated. For example, while grain boundaries have been known to affect PSC performance,[3] we are now setting out to resolve how the exact grain boundary characters influence PSCs. Also, the influences of stacking faults and dislocations on PSC performance have been much less explored due to the difficulty of identifying their existence and locations, but now with this important, exciting research, a new direction of study has become possible. Furthermore, the acquisition of the accurate atomic structure of HPs based on TEM will largely benefit first-principle modeling and enable more reliable coupled theoretical experimental studies.

This inspiring step represents the possibility of atomic-scale characterization of HPs through regular STEM systems

without specific, high-end facilities such as cryogenic stages. Rothmann et al.[7] further point out that the cryogenic conditions provide little noticeable contribution to reduce the beam sensitivity of the FAPbI$_3$ thin films. This is consistent with a recent study by Gao and coworkers.[12] The authors further claim that low electron accelerating voltage (80 kV) will lead to faster damaging of HPs rather than high accelerating voltage (200 and 300 kV), which suggests that the sacrifice on resolution for a structural stability during TEM observation with reduced electron energy may not be necessary. A conclusion can be thus drawn that the decisive factor for a successful atomic-scale STEM imaging of HPs is the dose rate. This finding points out a valuable research direction to perform in-depth STEM investigations of HPs and PSCs. For example, differential phase contrast imaging may help reveal the details of light atoms for the accurate determination of loss and migration of FA$^+$ ions, which remains to be explored. Electron energy loss spectroscopy can be used to explore the physical properties of grain boundaries, stacking faults, dislocations, and point defects. Furthermore, *in situ* measurements may be feasible for direct, atomic-scale observations of the structural evolution of HPs under external stimuli.[4,13] The adoption of higher efficiency detectors with better signal-to-noise ratios is also likely to improve the STEM imaging quality for HPs.

Seeing is believing. The direct observation of atomic-scale structures and defects under AC-STEM will lay a solid foundation for further exploring emerging materials sciences in the HP system, opening the door to discussing the exact relationships between materials properties, device performance, and those "invisible" yet key structures. This will no doubt benefit the technological development of PSCs and other perovskite electronics such as light-emitting devices and radiation detectors. In a broader context, soft crystalline materials like HPs are finding

prominent applications in energy technologies because they simultaneously exhibit high structural flexibility and tunable electronic-ionic properties. The research methodology demonstrated by Rothmann et al.[7] should be generic to studying all these promising materials, showing far-reaching impacts on the development of variable energy technologies and potentially influencing the renewable energy landscape of the future.

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