A Report on "Perovskite Solar Cells with Atomically Coherent Interlayers on SnO2 Electrodes" by Min et al. (2021)

Reviewer 2

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> I am wiser than this person; for it is likely that neither of us knows anything fine and good, but he thinks he knows something when he does not know it, whereas I, just as I do not know, do not think I know, either. I seem, then, to be wiser than him in this small way, at least: that what I do not know, I do not think I know, either.
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> Plato, *The Apology of Socrates*, 21d

> To err is human. All human knowledge is fallible and therefore uncertain. It follows that we must distinguish sharply between truth and certainty. That to err is human means not only that we must constantly struggle against error, but also that, even when we have taken the greatest care, we cannot be completely certain that we have not made a mistake.
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> Karl Popper, 'Knowledge and the Shaping of Reality'

Overview

Citation: Min, H., Lee, D. Y., Kim, J., Kim, G., Lee, K. S., Kim, J., Paik, M. J., Kim, Y. K., Kim, K. S., Kim, M. G., Shin, T. J., & Seok, S. I. (2021). Perovskite solar cells with atomically coherent interlayers on SnO2 electrodes. *Nature*, Vol. 598, No. 7881, pp. 444–450.

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Abstract Summary: This article reports on the formation of an atomically coherent interlayer between a SnO2 electron-transporting layer and a halide perovskite light-absorbing layer in perovskite solar cells. This interlayer, formed by coupling Cl-bonded SnO2 with a Cl-containing perovskite precursor, significantly enhances charge extraction and transport while reducing interfacial defects, leading to high power conversion efficiency and improved stability.

Key Methodology: The study utilized time-of-flight secondary ion mass spectrometry (ToF-SIMS), grazing-incident X-ray absorption fine structure (XAFS), synchrotron-based grazing-incidence wide-angle X-ray diffraction (GI-WAXD), high-resolution transmission electron microscopy (HR-TEM), and density functional theory (DFT) calculations to investigate the interlayer formation and its properties.

Research Question: Can an atomically coherent interlayer be formed between SnO2 electron-transporting layers and halide perovskite light-absorbing layers to improve the efficiency and stability of perovskite solar cells?

Summary

Is It Credible?

Min et al. claim to have developed perovskite solar cells with a remarkable power conversion efficiency of 25.8% (certified at 25.5%) and excellent operational stability, maintaining approximately 90% of their initial efficiency after 500 hours of continuous light exposure. The authors attribute these performance breakthroughs to the formation of an "atomically coherent interlayer" between the $SnO_{\square}$ electron-transporting layer and the perovskite light-absorbing layer. According to the article, this interlayer is achieved by combining a Cl-bonded $SnO_{\square}$ layer with a Cl-containing perovskite precursor, which enhances charge extraction and minimizes interfacial defects (p. 444).

The authors provide extensive spectroscopic and microscopic evidence to support the presence of a new interfacial phase, tentatively identified as $FASnCl_{\square}$. However, the central concept of an "atomically coherent" interlayer may overstate the definitive evidence. The authors acknowledge that the exact crystal structure of this layer is "not fully defined" (p. 447). Furthermore, there is a notable disconnect between their theoretical modeling and experimental observations. Density functional theory calculations are used to demonstrate the possibility of a stable cubic $FASnCl_{\square}$ interlayer, but the experimental grazing-incidence wide-angle X-ray diffraction data is inconsistent with a cubic structure, leading the authors to infer a tetragonal structure instead (pp. 446–447). While the authors suggest that a lower-symmetry tetragonal structure is reasonable for a thin film connecting two heterostructures, the discrepancy between the theoretical justification (which models a cubic structure) and the physical observation is not fully resolved through updated modeling.

Definitively proving that the structural coherence of this layer is the primary driver of the performance gains is also challenging. The high concentration of chloride

ions at the interface could independently passivate defects in the SnO□ or the perovskite, meaning the observed FASnCl□ layer might be a byproduct of these chemical conditions rather than the sole causal agent. This ambiguity is compounded by a missing device-level control experiment. While the authors use X-ray absorption fine structure analysis to examine the combination of Cl-bonded SnO□ with a Cl-free perovskite precursor, they do not report the full solar cell efficiency for this specific combination (p. 446). Without this control, it is difficult to isolate the individual contributions of the two Cl-containing components to the final device performance.

The reported efficiencies are highly credible and supported by independent certification, representing a major achievement. The authors attribute these gains to reduced interfacial defects, citing thermally stimulated current measurements (p. 448). However, because this technique measures total trap density across the entire device stack, assuming the observed defect reduction is exclusively interfacial may be overly definitive. Finally, while the long-term stability results are impressive, the 500-hour operational stability data appears to be based on a single champion device (p. 449). Without statistical data on the stability of multiple devices—unlike the efficiency data, which includes a robust sample size of 51 devices for the primary condition—the generalizability of this specific stability metric remains somewhat uncertain. Overall, the study presents a highly effective fabrication strategy that yields state-of-the-art perovskite solar cells, even if the precise structural nature and exact causal mechanisms of the interfacial layer require further elucidation.

The Bottom Line

Min et al. present a highly successful fabrication strategy for perovskite solar cells, achieving an impressive and independently certified power conversion efficiency of 25.5%. The core claim that these gains are driven by an "atomically coherent interlayer" is supported by substantial material characterization, though the exact crystal

structure and the specific causal mechanisms remain somewhat ambiguous. While the theoretical models and experimental data show some structural inconsistencies (though the authors offer a physical rationale for this), and the stability claims rely on a single champion device, the overall performance improvements represent a significant and credible contribution to photovoltaic research.

Potential Issues

Causal attribution of performance gains: The study establishes a strong correlation between a specific fabrication recipe, the spectroscopic signatures of an interfacial layer, and high device performance, but the causal claim that the "atomically coherent" nature of this interlayer is the primary reason for the improvements may not be definitively proven (pp. 444–445). An alternative explanation is that the chemical conditions required to form the layer—specifically, the high concentration of chloride ions at the interface—could independently cause beneficial effects, such as passivating defects in the SnO□ or the perovskite. The observed FASnCl□ layer might be a byproduct of these direct passivation mechanisms rather than the primary causal agent itself. The research design does not appear to fully deconvolve the effect of the interlayer's specific structure from the general chemical effects of the precursors used to create it.

Ambiguity of the central concept: The article's central claim rests on the formation of an "atomically coherent interlayer," but this term is not precisely defined and may overstate the evidence. While the authors provide multi-modal evidence for chemical bonds across the interface (Sn-Cl), the term "atomically coherent" implies a near-perfect, defect-free, single-crystal-like junction. This strong claim is somewhat undermined by the authors' own admission that the interlayer's crystal structure is "not fully defined" (p. 447). Furthermore, the High-Resolution Transmission Electron Microscopy (HR-TEM) image, interpreted as showing a seamless interface where "boundaries could not be distinguished," demonstrates good physical contact but does not by itself prove atomic-level coherence across the entire interface (p. 447). The term may function more as a conceptual model than a rigorously proven physical state.

Inconsistency between theoretical model and experimental findings: The article presents Density Functional Theory (DFT) calculations that "demonstrate the possi-

bility of generating a very stable FASnCl□ interlayer" with a cubic structure (p. 446). However, the primary experimental evidence from Grazing-Incidence Wide-Angle X-ray Diffraction (GI-WAXD) directly contradicts this model. The authors state that "the results of GI-WAXD are inconsistent with the cubic structure" and instead infer a tetragonal structure from their data (p. 447). This creates a disconnect where the theoretical model used to justify the energetic favorability of the interlayer describes a different structure from the one observed experimentally. The authors do offer a physical rationale for this, suggesting that "given that the interlayer is a thin film that connects the two heterostructures between the perovskite and SnO□, it may be more reasonable for the lattice to have a tetragonal structure with a lower symmetry than that of a cubic structure" (p. 447). However, the theoretical model itself is not updated to reflect this physical reality.

Missing device-level control experiments: The study's experimental design may be missing a key control device needed to fully isolate the contributions of the two main components: the Cl-bonded SnO□ (Cl-bSO) and the Cl-containing perovskite precursor (Cl-cPP). While the article analyzes the combination of Cl-bSO with a Cl-free perovskite precursor using X-ray absorption spectroscopy (XAFS) to show that the Sn-Cl bond count does not increase, it does not report the performance of a full solar cell fabricated with this combination (p. 446). Without this device-level data, it is difficult to determine whether the Cl-bSO provides any benefit on its own or if the performance gains stem entirely from the Cl-cPP. This missing control makes it harder to parse the individual versus combined effects of the novel components on final device efficiency and stability.

Interpretation of defect measurements: The article attributes the improved performance to a reduction in interfacial defects, primarily citing Thermally Stimulated Current (TSC) measurements that show a lower total trap density in the Cl-bSO device compared to a TiO□ control (p. 448). However, TSC is a bulk measurement technique that quantifies the total density of electronic traps throughout the entire device

stack and cannot spatially resolve their location. The conclusion that the reduction is interfacial rests on the assumption that "the difference between the PSC prepared with planar Cl-bSO and a TiO□ ETL is due to only the interface" (p. 448). This assumption may be too strong, as the properties of the underlying electron transport layer are known to influence the crystallization and bulk defect density of the perovskite film deposited on top of it. Therefore, it is possible that some of the observed reduction in defects occurs in the perovskite bulk rather than being confined exclusively to the interface.

Uncertainty in the interlayer's crystal structure: The article infers a specific tetragonal crystal structure for the ~2 nm thick interlayer, providing lattice parameters based on new diffraction spots observed in GI-WAXD patterns (p. 447). However, determining a complete crystal structure for a thin, buried layer from a limited number of faint and elongated diffraction spots is a significant challenge. The authors themselves acknowledge this uncertainty by stating, "we label the interlayer as FASnCl□ rather than FASnCl□, because it is not fully defined" (p. 447). While presenting the tetragonal structure as the most plausible hypothesis is reasonable, the evidence may not be sufficient to consider the structure definitively solved.

Generalizability of stability claims: The article's impressive long-term stability result—maintaining 90% of initial efficiency after 500 hours of continuous operation—is based on a single "best-performing" device (p. 449, Fig. 4d). While reporting the performance of a champion cell is standard practice, the lack of statistical data on the stability of multiple devices limits the generalizability of this key claim. It is unclear from the data presented whether this level of stability is typical for devices made with this method or if the champion device is an outlier. The efficiency data is better supported by a statistical distribution of 51 devices (Fig. 4a), but similar data for stability is not provided.

Minor methodological and presentation issues: Several minor issues related to transparency and clarity are present in the article. The current density-voltage (J-

V) curves in Figure 4c are presented, but the Methods section does not specify the voltage scan rate, a key parameter for perovskite solar cells that can be subject to hysteresis. Additionally, the statistical comparison in Figure 4a is based on imbalanced sample sizes, with the main group having N=51 while control groups have much smaller sizes (N=11 to 16), which reduces the statistical power of the comparison.

Future Research

Isolating chemical versus structural effects: Future research should include comprehensive device-level control experiments to separate the effects of chemical passivation from structural coherence. Fabricating and testing full solar cells using Cl-bonded SnO□ with Cl-free perovskite precursors, and comparing them directly to the combined Cl-system, would help determine whether the performance gains stem primarily from the presence of chloride ions or the specific formation of the FASnCl□ interlayer.

Resolving the interfacial crystal structure: Further investigation is needed to fully reconcile the theoretical models with the experimental observations of the interlayer's crystal structure. While the authors propose a physical rationale for the observed symmetry lowering, advanced characterization techniques, such as cross-sectional scanning transmission electron microscopy coupled with localized elemental mapping, could be used alongside updated density functional theory calculations that explicitly model the experimentally inferred tetragonal structure.

Statistical validation of long-term stability: To strengthen the claims regarding operational stability, future studies should report maximum power point tracking data across a statistically significant sample size of devices. Providing distribution metrics for stability, similar to those provided for initial power conversion efficiency, would confirm whether the impressive 500-hour stability observed in the champion device is consistently reproducible across standard fabrication batches.

Spatially resolved defect characterization: Because thermally stimulated current measurements capture bulk trap densities, subsequent research could employ spatially resolved techniques to definitively locate the defect reductions. Methods such as depth-resolved deep-level transient spectroscopy or cross-sectional Kelvin probe force microscopy would help confirm whether the defect mitigation is truly confined

to the interface or if it extends into the bulk perovskite layer.

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