Multiscale Modeling of Perovskite Solar Cells: A Review of DFT, TMM, and SCAPS-1D Approaches for Performance Optimization and Degradation Analysis

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INTRODUCTION

Perovskite solar cells were introduced in 2009 at 3.8% efficiency [1] and reached 27.3% by 2024 [2]. Despite this progress, the gap between laboratory champions and stable modules persists. PSC performance is governed by three physical phenomena: defect formation and ion migration at the atomic scale, coherent optical interference in nanometer-thin stacks, and drift-diffusion carrier transport at the device scale — each requiring DFT, TMM, and SCAPS-1D, respectively. A critical unresolved problem is that these frameworks are applied in isolation, producing cascading systematic errors that this review quantifies and resolves.

DFT: ATOMISTIC FOUNDATION

Kohn-Sham DFT accuracy is dominated by the exchange-correlation functional. For $MAPbI_3$, GGA-PBE severely underestimates Eg due to self-interaction error; SOC inclusion further reduces Eg by ~0.4 eV from Pb 6p splitting [3]. HSE06+SOC achieves ±0.05 eV agreement with experiment and is mandatory for device simulation. Defect formation enthalpy ΔHf classifies intrinsic defects as shallow (VMA, VI) or deep traps (Ii, PbI at 0.6–0.8 eV from CBM) [4]. NEB barriers Ea(VI) ~ 0.1–0.3 eV and Ea(VMA) ~ 0.4–0.6 eV set J-V hysteresis timescales [5,6]. Table 1 summarizes functional performance.

Table 1. DFT functional comparison for $MAPbI_3$ bandgap calculation.

Functional	Eg (eV)	SOC	Deviation	Recommendation
GGA-PBE	1.20–1.40	No	−0.20 to −0.40 eV	Not recommended for device simulation
GGA-PBE+SOC	0.50–0.80	Yes	−0.80 to −1.10 eV	Overcorrects — not suitable
HSE06	1.45–1.55	No	−0.05 to −0.15 eV	Acceptable if SOC unavailable
HSE06+SOC	1.55–1.65	Yes	±0.05 eV	Recommended

TRANSFER MATRIX METHOD: OPTICAL MODELING

TMM computes the complex electric field profile $|E(x,\lambda)|^2$ through the full multilayer stack via 2×2 transfer matrices, yielding the volumetric generation rate $G(x,\lambda)$. For the canonical FTO/TiO_2/$MAPbI_3$(400 nm)/Spiro/Au device, TMM gives Jsc ~ 21.5 mA/cm² vs. Beer–Lambert's ~19.8 mA/cm² — an 8.6% systematic error from constructive interference near the Au back contact [7]. This Jsc discrepancy propagates directly into SCAPS-1D PCE predictions (+1.5–2.0% absolute). Table 2 compares the two approaches quantitatively.

Table 2. TMM vs. Beer–Lambert: quantitative comparison for 400 nm $MAPbI_3$ absorber

Parameter	Beer–Lambert	TMM	Impact
Jsc (400 nm absorber)	~19.8 mA/cm²	~21.5 mA/cm²	+8.6% systematic error
Optical interference	Ignored	Fully accounted	Critical for <500 nm
PCE overestimation	+1.5–2.0% abs.	Reference	Propagates to Voc, FF
Applicability	Thick absorbers >1 µm	All thin-film PSC stacks	TMM mandatory for PSC

SCAPS-1D: DEVICE-SCALE SIMULATION

SCAPS-1D self-consistently solves the Poisson and continuity equations under AM1.5G illumination [8], including SRH, radiative ($B \sim 10^{-10}$ cm³/s), and Auger recombination. The primary reproducibility barrier is the two-orders-of-magnitude spread in reported trap densities Nt = 10^{15}–10^{17} cm^{-3} [9]. Parameter sensitivity: ΔEg = 0.1 eV → ΔPCE ~1.5%; two-decade variation in Nt → ΔPCE ~3–5%. Table 3 provides a parameter sensitivity ranking essential for reliable simulation.

Table 3. SCAPS-1D parameter sensitivity ranking for PSC performance.

Parameter	Reported range	Sensitivity to PCE	Impact on Voc	Priority
Bandgap Eg	1.55–1.65 eV	ΔEg=0.1 eV → ΔPCE ~1.5%	ΔVoc ~80 mV	Critical
Trap density Nt	10^{15}–10^{17} cm^{-3}	2 decades → ΔPCE ~3–5%	~150 mV	Critical
Interface S (rec. velocity)	10^2–10^5 cm/s	FF < 0.70 at S > 10^4	~100–200 mV	High
Hole mobility µp	10–20 cm²/Vs	Low sensitivity	<30 mV	Medium
Carrier lifetime τ	1–100 ns	Low sensitivity at τ > 10 ns	<20 mV	Low

COMSOL: COUPLED DEGRADATION MODELING

COMSOL Multiphysics extends analysis into 2D/3D time-dependent domains coupling thermal ($\nabla\cdot(\kappa\nabla T) = Q$), mechanical ($\nabla\cdot\sigma + F = 0$), and Nernst-Planck ion transport. It uniquely resolves: (i) J-V hysteresis from halide redistribution (τion ~ 1–100 s); (ii) thermal degradation via Arrhenius kinetics (Ea ~ 0.9–1.2 eV for $MAPbI_3$); and (iii) CTE-mismatch interfacial stress (>50 MPa after thermal cycling). Simulated PCE loss under ISOS-D-2 (85°C/85%RH) is ~0.5%/day.

MULTISCALE INTEGRATION AND PARAMETER TRANSFER HIERARCHY

The three scales are rarely coupled consistently. GGA-PBE Eg used directly in SCAPS-1D causes systematic Voc underestimation of 120–200 mV; Beer–Lambert in place of TMM introduces ΔJsc ~ 1.5–2.0 mA/cm² and ΔPCE ~ 1.5–2.5% absolute. Combined, uncoupled errors exceed ±3% PCE absolute. The proposed hierarchy (Table 4, Fig. 1) enforces explicit uncertainty propagation at each interface and is expected to reduce systematic error to below 1% absolute (see Fig. 2). Validation must extend beyond J-V curves to EQE (optical accuracy), TRPL (recombination dynamics), and EIS (interface traps).

Table 4. Proposed DFT→TMM→SCAPS-1D→COMSOL parameter transfer hierarchy.

Level	Tool	Key Outputs	Passed to	Uncertainty
1	DFT (HSE06+SOC)	Eg, χ, m, εr, ε(ω), Et, Ea	TMM + SCAPS-1D	ΔEg ≈ ±0.05 eV
2	TMM	G(x,λ), Jsc, R(λ), A(λ)	SCAPS-1D	ΔJsc ≈ ±1.5 mA/cm²
3	SCAPS-1D	J-V, EQE, Nt, S, τSRH	COMSOL	ΔVoc ≈ ±40 mV
4	COMSOL	PCE(t), stress, c(x,y,t)	Stability prediction	ΔPCE ≈ ±1.5%

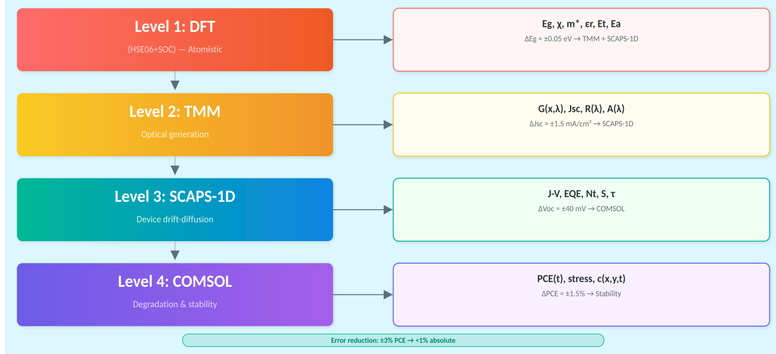

Fig. 1. Proposed DFT→TMM→SCAPS-1D→COMSOL parameter transfer hierarchy with uncertainty propagation.

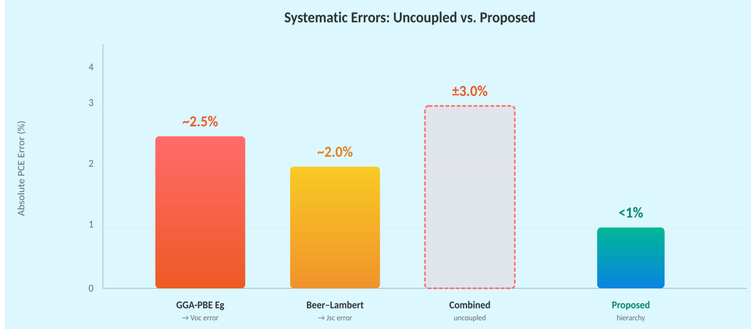

Fig. 2. Systematic PCE errors from uncoupled vs. proposed hierarchy.

CONCLUSIONS

This review identifies three systematic deficiencies in current PSC simulation practice and proposes concrete solutions.

1. DFT: HSE06+SOC is mandatory — GGA-PBE underestimates Eg by 0.15–0.25 eV, introducing ~160 mV systematic Voc error.
2. TMM: Beer–Lambert introduces ~8.6% Jsc error for 400 nm absorbers; TMM-derived $G(x,\lambda)$ is mandatory for PSC simulation.
3. SCAPS-1D: Two-order-of-magnitude variability in Nt, compounded by uncorrected DFT inputs, is the primary reproducibility barrier. The proposed hierarchy reduces systematic PCE prediction error from ~3% to below 1% absolute. Physics-informed neural networks and digital twin architectures represent the next generation of PSC simulation but require this scale-bridging consistency as a prerequisite.

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