

# Nanoporous GaN/*n*-type GaN: A Cathode Structure for ITO-Free Perovskite Solar Cells

Kwang Jae Lee,<sup>+</sup> Jung-Wook Min,<sup>+</sup> Bekir Turedi, Abdullah Y. Alsalloum, Jung-Hong Min, Yeong Jae Kim, Young Jin Yoo, Semi Oh, Namchul Cho, Ram Chandra Subedi, Somak Mitra, Sang Eun Yoon, Jong H. Kim, Kwangwook Park, Tae-Hoon Chung, Sung Hoon Jung, Jong H. Baek, Young Min Song, Iman S. Roqan, Tien Khee Ng, Boon S. Ooi,\* and Osman M. Bakr\*



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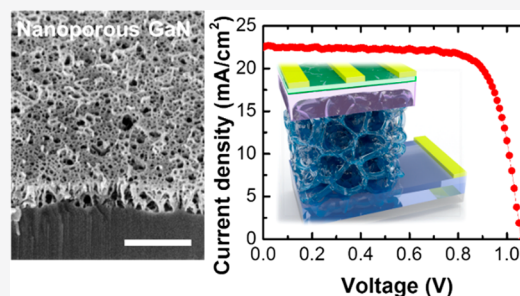


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**ABSTRACT:** Introducing suitable electron/hole transport layers and transparent conductive layers (TCLs) into perovskite solar cells (PSCs) is key to enhancing the selective extraction of charge carriers and reducing surface recombination losses. Here, we introduce nanoporous gallium nitride (NP GaN)/*n*-type GaN (*n*-GaN) as a dual-function cathode structure for PSCs, acting as both the TCL and the electron transport layer (ETL). We demonstrate that the hierarchical NP GaN structure provides an expanded interfacial contact area with the perovskite absorber, while the *n*-GaN under the NP GaN displays high transmittance in the visible spectrum as well as lateral electric conductivity higher than that of a conventional ITO film. Prototype MAPbI<sub>3</sub> PSCs based on this NP GaN/*n*-GaN cathode structure (without an extra ETL) show a power conversion efficiency of up to 18.79%. The NP GaN/*n*-GaN platform demonstrated herein paves the way for PSCs to take advantage of the widely available heterostructures of mature III-nitride-based technologies.



Halide perovskites with the general formula AMX<sub>3</sub> (where A = CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>, CH(NH<sub>2</sub>)<sub>2</sub><sup>+</sup>, Cs<sup>+</sup>; M = Pb<sup>2+</sup>, Sn<sup>2+</sup>; X = I<sup>-</sup>, Br<sup>-</sup>, Cl<sup>-</sup>) have surged to the forefront of optoelectronic materials primarily because of the outstanding performance of perovskite solar cells (PSCs).<sup>1–8</sup> Bandgap tunability (1.5–3.1 eV), a high absorption coefficient (10<sup>4</sup>–10<sup>5</sup> cm<sup>-1</sup>), low exciton binding energy (<50 meV), and simplicity of fabrication are some of the few properties that played a key role in cementing the place of halide perovskites among solar cell materials.<sup>9–14</sup> However, because dopant–host interactions in perovskites are distinct from those of traditional semiconductor materials (for example, Si:B, GaAs:Mg, and AlN:Si),<sup>15–17</sup> perovskites have not yet been formed into *p-i-n* junctions through doping. Thus, heterogeneous materials need to be integrated to create junctions that promote the charge extraction and collection in PSCs. In this respect, one of the critical issues concerning the fabrication of PSCs is designing the most suitable cathode structure by combining key material components for the extraction of photogenerated electrons, such as an electron transport layer (ETL) and a transparent conductive layer (TCL, cathode electrode).

In PSC cathodes, the ETL is specially designed not only to extract photocurrent but also to exhibit chemical and physical properties commensurate with those of the rest of the PSC structure, such as chemical durability, interfacial properties,

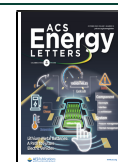
thermal/electrical conductivities, bandgap alignment, and optical properties (e.g., refractive index and transmittance).<sup>18–20</sup> TiO<sub>2</sub>,<sup>19</sup> ZnO,<sup>20</sup> SnO<sub>2</sub>,<sup>21</sup> and SrSnO<sub>3</sub><sup>22</sup> are the best-known starting materials for forming ETLs; however, these wide-bandgap oxide materials have several disadvantages, which may include poor electron mobility,<sup>19–22</sup> chemical instability in acid/base solutions,<sup>23</sup> reactivity with the perovskite layer,<sup>23</sup> and high-temperature processing,<sup>24,25</sup> as well as difficulty in tuning band structure, particle size, and film morphology.<sup>26</sup>

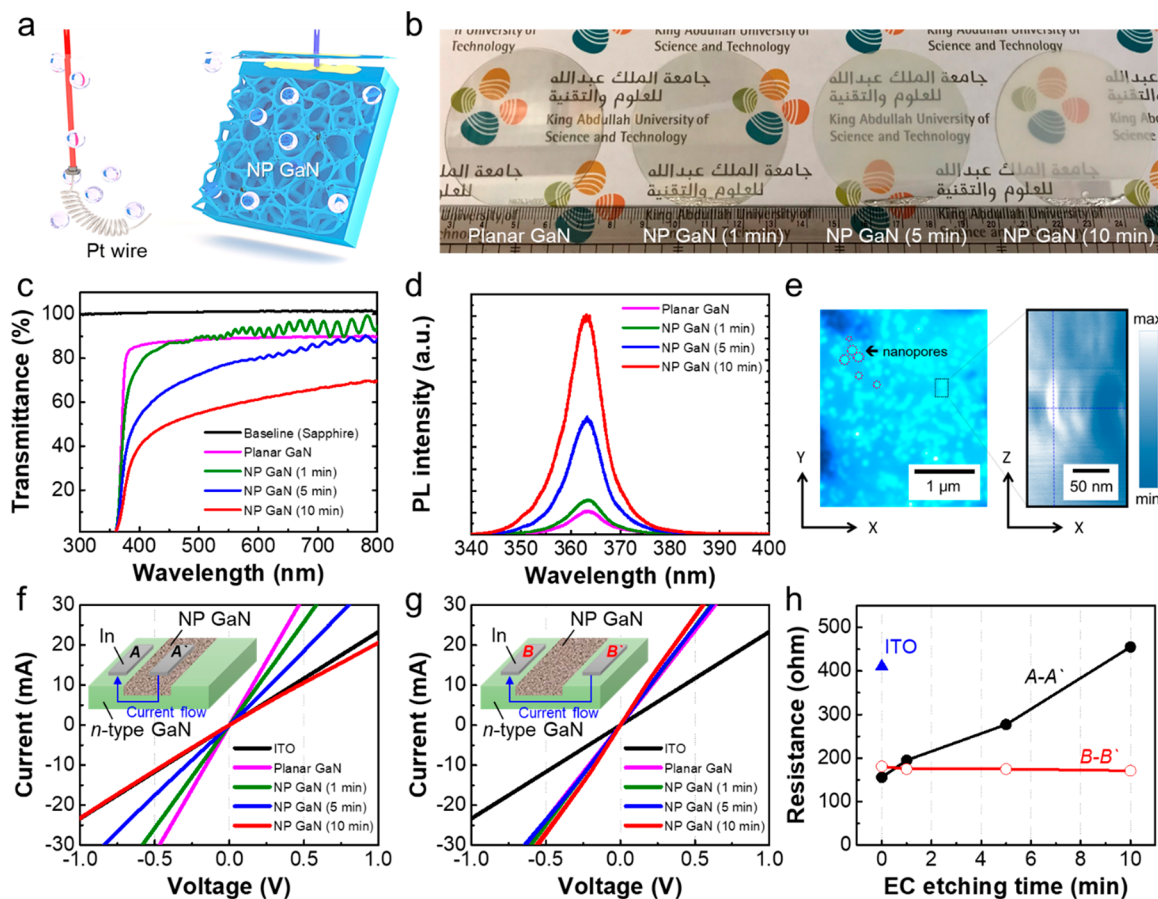
Besides the ETL, a TCL such as indium tin oxide (ITO) is an essential part of a PSC for extracting photogenerated charge carriers from the perovskite solar absorber.<sup>2,27,28</sup> The two requirements for a suitable TCL are high transmittance, to deliver more light to the perovskite absorber, and high electrical conductivity, to extract photogenerated electrons more efficiently through the horizontal current path.<sup>28</sup>

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**Figure 1.** (a) Schematic diagram of electrochemical etching process for NP GaN. (b) Photographs of NP GaN obtained at various electrochemical etching times. (c) Transmittance of NP GaN as a function of electrochemical etching time. (d) PL spectra of NP GaN as a function of electrochemical etching time. (e) Confocal PL images of photons generated by nanopore-induced Rayleigh scattering. Current–voltage curve (f) through the NP GaN region; here the current flow through NP GaN is denoted as A–A', (g) lateral electrical transport under the NP GaN layer (denoted as B–B'), and (h) resistances of NP GaN as a function of electrochemical etching time.

Unfortunately, during the past decade, the price of ITO has continuously risen because of the scarcity of indium, which may be depleted within the next decade.<sup>29–31</sup>

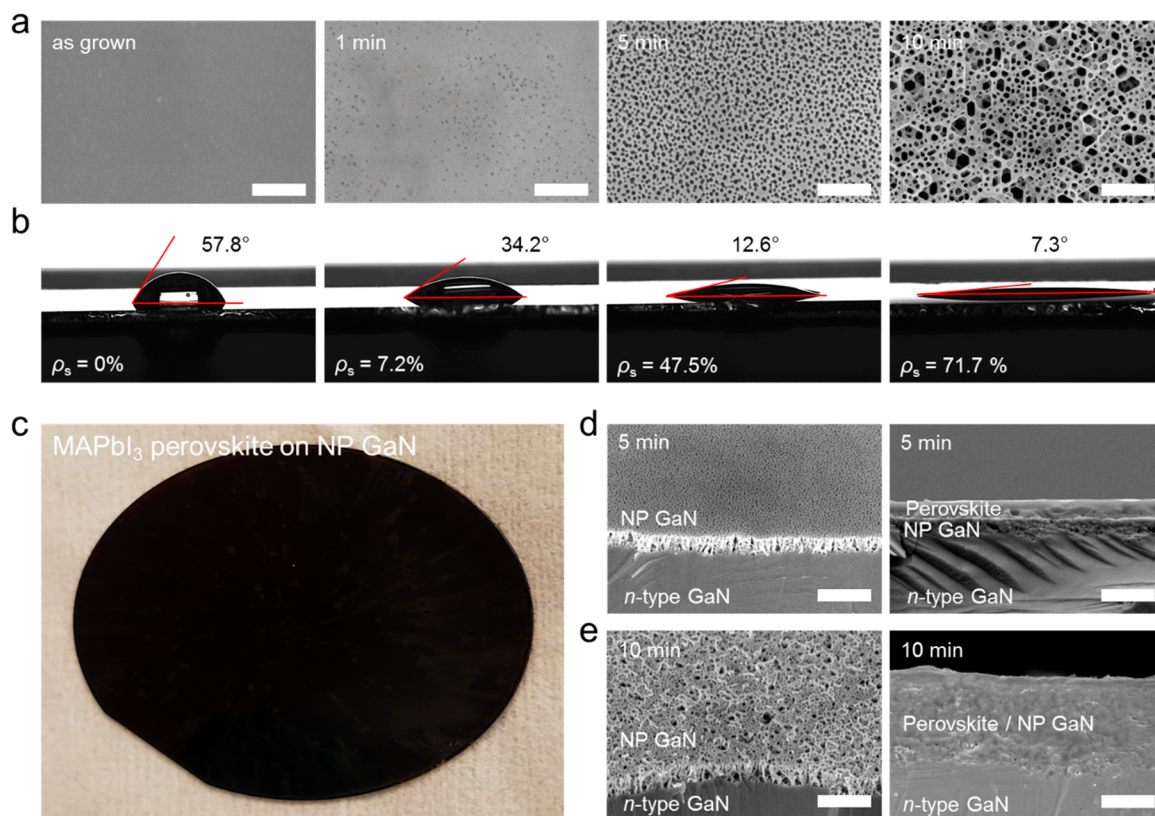
Gallium nitride (GaN) possesses excellent optical and electrical properties because of its direct bandgap ( $E_g \approx 3.4$  eV), transmittance exceeding 82% over the entire visible wavelengths, and high electron mobility ( $\sim 1000$  cm<sup>2</sup>/(V s)).<sup>32</sup> Facile control over the doping type at the desired carrier concentrations and the ease of  $E_g$  tuning with alloys (Ga<sub>x</sub>Al<sub>y</sub>In<sub>1-x-y</sub>N) allow for versatile tailoring of GaN's electrical and optical characteristics.<sup>33</sup> Furthermore, the thermal, mechanical, and chemical stability of GaN enable practical applications of light-emitting diodes (LEDs),<sup>34,35</sup> laser diodes (LDs),<sup>36</sup> and photodetectors (PDs).<sup>37</sup> These desired characteristics have attracted researchers to study the combination of GaN with perovskites for new concepts of device applications such as piezo-phototronics, photovoltaics, and nanostructured LDs.<sup>38–42</sup>

GaN is generally grown by metal–organic chemical vapor deposition (MOCVD),<sup>43</sup> hydride vapor phase epitaxy (HVPE),<sup>44</sup> molecular beam epitaxy (MBE),<sup>45</sup> or one of other analogous pathways.<sup>46,47</sup> The latest growth technologies have demonstrated their capacity for mass producing GaN wafers (>12 in.) with high throughput, low fabrication costs, and the ability to grow films not only on monocrystalline substrates but also on noncrystalline substrates such as glass

and flexible polymer substrates.<sup>48–50</sup> Compared to existing commercialized cathode structures, the price of GaN/substrate is competitive and is likely to decrease with further adoption (Table S1). While some studies have demonstrated potential advantages of GaN for perovskite technology, such as improved stability<sup>38</sup> or the use of GaN as an ETL material<sup>39–41</sup> in current device architectures, it is by eliminating undesirable heterogeneous interfaces, simplifying the device fabrication process, and expanding the design versatility of perovskite devices that GaN, arguably, could have the largest impact (by taking advantage of its versatile doping in combination with its monolithic growth processes).

Motivated by this goal, we introduce nanoporous (NP) GaN epitaxially grown on *n*-type GaN (*n*-GaN), referred to here as (NP GaN/*n*-GaN), as a dual-function cathode structure containing both the TCL and the ETL for spin-coated methylammonium lead iodide (MAPbI<sub>3</sub>) PSCs. Our finite-difference time-domain (FDTD) and rigorous coupled-wave analysis (RCWA) simulations show that this structure enhances photon harvesting in the cell because of the gradual change in the effective refractive index through the NP GaN/MAPbI<sub>3</sub> layer. While one would expect that the increased interfacial contact area between the NP GaN and the MAPbI<sub>3</sub> may also increase the probability that interface defects form, which could militate the device performance, a spontaneously forming Ga<sub>2</sub>O<sub>3</sub> layer on the surface of the NP GaN (during the





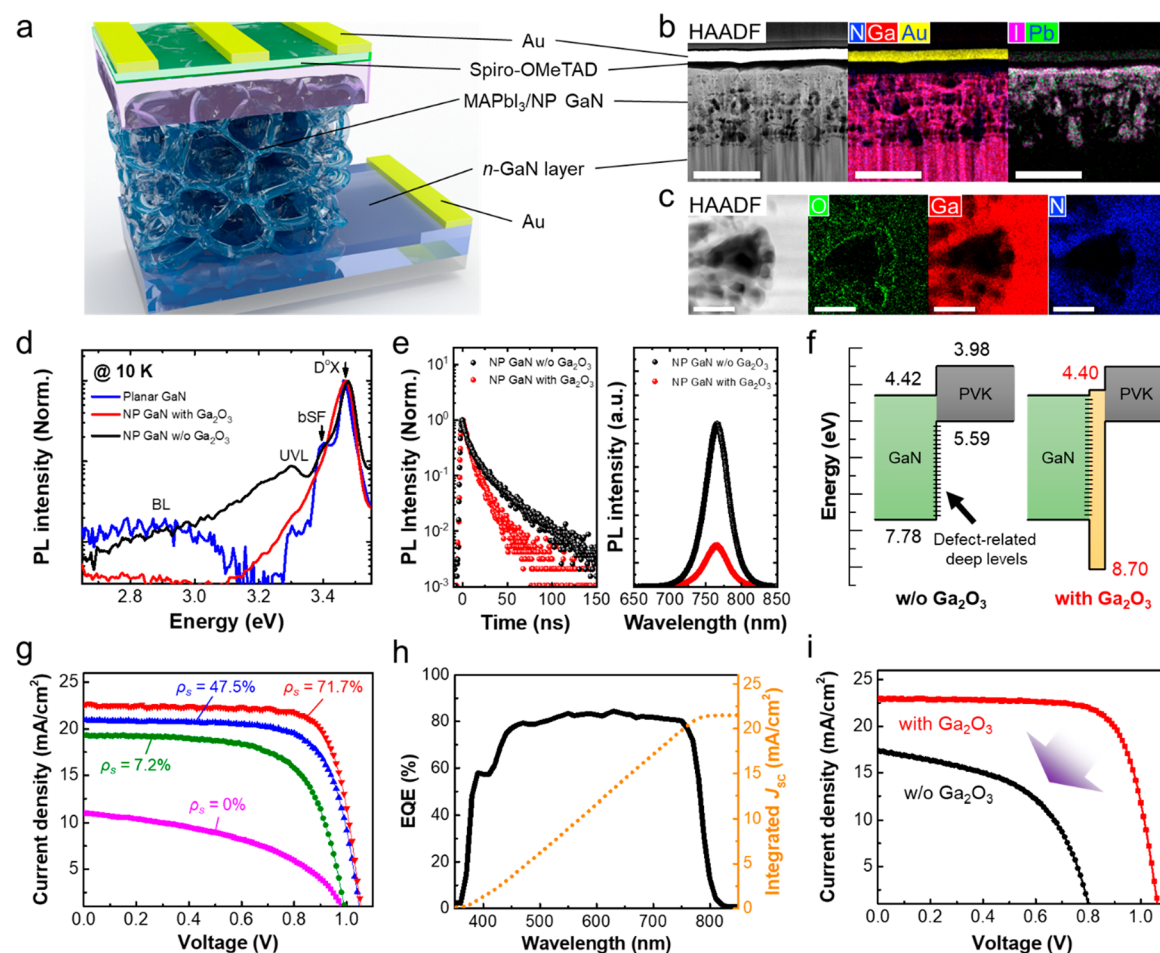
**Figure 2.** Plan-view scanning electron microscopy (SEM) images of (a) *n*-GaN layer template and NP GaN formed after 1, 5, and 10 min of electrochemical etching. Scale bar is 500 nm. (b) Water contact angle measurement for *n*-GaN template with NP GaN formed after 1, 5, and 10 min of electrochemical etching. (c) Photograph of perovskite MAPbI<sub>3</sub> spin-coated thin film on a 2 in. NP GaN wafer. Bird's-eye and cross-sectional SEM images of (d) NP GaN formed after 5 min and (e) 10 min of electrochemical etching. Scale bar is 1.0  $\mu$ m.

electrochemical etching-fabrication of the NP GaN layer) acts as an interlayer between the perovskite and NP GaN and, thereby, passivating defect-related deep levels. To illustrate these advantages, we demonstrate prototype MAPbI<sub>3</sub> PSCs based on this NP GaN/*n*-GaN cathode structure (without any other ETL) with promising power conversion efficiencies (PCEs) of up to 18.79%. Our work shows that III-nitride-based nanostructure can be versatile components in perovskite-based optoelectronics, enabling new device architectures that do away with rate-limiting material layers.

To fabricate NP GaN, the electrochemical etching of *n*-GaN was conducted as shown in Figure 1a. The 10  $\mu$ m-thick *n*-GaN thin films were grown on the double-polished sapphire substrate to ensure high transmittance to visible light. Figure 1b displays the 2 in. wafers of as-grown *n*-GaN and NP GaN formed by 1, 5, and 10 min of electrochemical etching at an applied bias of 12 V (Figure S1). Upon increasing the etching time, the transmittance of GaN (Figure 1c) was systematically decreased, and the integrated photoluminescence (PL) intensity (Figure 1d) was increased accordingly, because of the scattering effect from the formed nanopores. Figure 1e shows optical and confocal microscopy images of nanopores under a luminous source (450 nm). Photons penetrating the NP GaN interact with the nanopores ( $D_{\text{avg}} = 37.5$  nm), leading to Rayleigh scattering. This optical property of NP GaN causes the trajectory of photons to fluctuate, contributing to the enhancement in the light harvesting of the NP GaN medium. In terms of electrical properties, the current flow through NP GaN (denoted as A–A') was influenced by etching; the resistance was increased by increasing the etching time for the

NP GaN structure (Figure 1f) because of the pore volume fraction, including pore interconnection and tortuosity.<sup>51</sup> On the other hand, lateral electrical transport through the *n*-GaN/*n*-GaN structure (denoted as B–B') showed nearly the same resistivity regardless of the etching time (Figure 1g). Despite the presence of a porous layer along the direction of current flow, the NP GaN structure exhibited a lateral electric conductivity higher than that of a conventional ITO film (Figure 1h). Thus, the NP GaN/*n*-GaN structure serves as a dual-function layer exhibiting properties of both a TCL and an ETL.

To explore the possibility of depositing a perovskite film on NP GaN by spin-coating of a perovskite solution, we conducted contact angle measurements. We calculated the surface area fraction ( $\rho_s$ ) of NP GaN using plan-view scanning electron microscopy (SEM) images (Figure 2a). Figure 2b shows the contact angles measured for deionized water as a function of  $\rho_s$ . The hydrophobicity of the dense *n*-GaN film was indicated by a large contact angle of 57.8°. Increasing the value of  $\rho_s$  of NP GaN caused the contact angle to decrease (Figure S2), with angles of 34.2°, 12.6°, and 7.3° measured at  $\rho_s$  values of 7.2%, 47.5%, and 71.7%, respectively; this trend clearly reflects the hydrophilicity induced by introducing nanopores into the GaN surface. This result is attributed to the surface roughness and oxidation of the GaN surface to Ga<sub>2</sub>O<sub>3</sub> (or GaO<sub>3</sub><sup>3-</sup>) obtained by electrochemical etching, which induces surface tension energy higher than that observed for an intrinsic dense *n*-GaN surface.<sup>52</sup> After ozone plasma treatment for 10 min, we spin-coated MAPbI<sub>3</sub> perovskite onto a 2 in. NP GaN wafer (Figure 2c). Figure 2d,e shows how the



**Figure 3.** (a) Schematic illustration of the PSC structure including NP GaN/MAPbI<sub>3</sub>. (b) Cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of PSC. Energy-dispersive X-ray spectroscopy (EDX) elemental mapping of N, Ga, and Au for the structural overview and of Pb and I confirm that those materials are well incorporated into NP GaN. Scale bar is 1  $\mu\text{m}$ . (c) Cross-sectional HAADF-STEM image of the NP GaN/planar GaN interface and EDX elemental mapping of O, Ga, and N, respectively. Scale bar is 100 nm. (d) Low-temperature (10 K) PL measurements of planar GaN template and NP GaN (with and without Ga<sub>2</sub>O<sub>3</sub>) samples for defect study. Comparison of NP GaN with and without Ga<sub>2</sub>O<sub>3</sub> by (e) normalized transient PL decay profile (left) and steady-state PL (right). (f) Comparison of energy band diagrams for devices with and without Ga<sub>2</sub>O<sub>3</sub>. (g) Current–voltage characteristics of MAPbI<sub>3</sub> PSCs with planar GaN layer and NP GaN structures with porosities of  $\rho_s = 0, 7.2, 47.5,$  and  $71.7\%$ . (h) Incident photon-to-current efficiency of solar cell using NP GaN ( $\rho_s = 71.7\%$ ) and (i) comparison of current–voltage characteristics for NP GaN/MAPbI<sub>3</sub> solar cells with and without Ga<sub>2</sub>O<sub>3</sub> interlayer.

morphology of the MAPbI<sub>3</sub> perovskite films varied with the  $\rho_s$  of NP GaN as a result of changes in the spin-coating conditions. At high porosities ( $\rho_s > 71.7\%$ ), the perovskite film was embedded in the NP GaN; however, at low porosities ( $\rho_s < 47.5\%$ ), the perovskite film was separated from the NP GaN. These results show that the perovskite could be confined in the nanopores when the  $\rho_s$  of the NP GaN layer was sufficiently high ( $\rho_s > 71.7\%$ ). To summarize, NP GaN exhibits the prerequisite optical, electrical, and structural characteristics to be a promising substitute for the current layers employed as ETLs/TCLs in PSC devices.

To confirm that the NP GaN structure can serve as an alternative to conventional ETLs and TCLs, we fabricated proof-of-concept MAPbI<sub>3</sub> PSCs based on this structure. Figure 3a shows a schematic illustration of a PSC featuring a NP GaN/MAPbI<sub>3</sub> heterostructure. Using planar *n*-GaN, NP GaN was formed by electrochemical etching for different durations. To identify the various layers of the solar cell structure, we cross-sectioned the structure by focused-ion-beam (FIB) processing and then probed the cross-section using trans-

mission electron microscopy (TEM). Figure 3b shows corresponding cross-sectional high-angle annular dark-field scanning transmission microscopy (HAADF-STEM) images. The various layers of the solar cell are visually distinct, and both the planar GaN layer and perovskite-coated NP GaN are clearly shown. In STEM mode, energy-dispersive X-ray spectroscopy (EDX) with element mapping of N, Ga, and Au confirmed that the NP GaN structure remained well intact after solar cell fabrication. Elemental mapping of I and Pb revealed that the perovskite material was well-incorporated deep into the NP GaN structure (Figure S3). Additional EDX mapping for other elements was also performed at the interface of the pristine NP GaN layer without perovskite (Figure 3c). Note that in the area around the interface between the NP GaN and planar GaN layers, the constituent elements (O, Ga, and N) are distributed uniformly throughout the topmost GaN surface of the NP GaN structure. Interestingly, a highly intense peak corresponding to O was found at the edge of the porous structures, suggesting that the Ga<sub>2</sub>O<sub>3</sub> interlayer was formed as an outer shell around the NP GaN. This Ga<sub>2</sub>O<sub>3</sub> likely resulted



from the oxalic acid used during the electrochemical etching of NP GaN.

To better understand the role of Ga<sub>2</sub>O<sub>3</sub> in NP GaN, we performed a series of optical characterizations on NP GaN samples (with and without Ga<sub>2</sub>O<sub>3</sub>). To fabricate NP GaN without Ga<sub>2</sub>O<sub>3</sub>, the sample was cleaned with acetone and isopropanol (IPA) and then rinsed with deionized water in advance. Additional oxides were removed by dipping the sample in buffered oxide etch (BOE) for 5 min. Using a planar GaN layer as a reference, NP GaN samples were examined by low-temperature (10 K) PL measurements to confirm the crystallinity of GaN and analyze several defect states of the NP structures. Figure 3d shows neutral donor bound exciton (*D*<sup>0</sup>X) peaks around 3.478 eV detected in three samples, which represent the dominant emission spectrum of GaN. These peaks agree well with the *D*<sup>0</sup>X peaks reported previously for MOCVD planar GaN layer templates grown on a sapphire substrate.<sup>52</sup> It should be noted that *D*<sup>0</sup>X peaks can be observed only in highly crystalline homoepitaxial GaN samples; thus, the electrochemical etching process used to form NP GaN does not degrade the crystal quality of GaN.<sup>53,54</sup> It should also be noted that the PL spectrum of the planar GaN layer sample and that of the NP GaN sample without Ga<sub>2</sub>O<sub>3</sub> displayed several interesting features. For instance, a secondary peak around the dominant peak (*D*<sup>0</sup>X ≈ 3.4 eV), attributed to a basal-plane stacking fault (bSF), was observed in the two samples.<sup>55</sup> The bSF emission peak is a well-known intrinsic defect peak and is commonly observed in epitaxially grown GaN. Other defect-related transition lines of ultraviolet luminescence (UVL) near 3.3 eV and blue luminescence (BL) around 2.9–3.0 eV were also observed.<sup>56–58</sup> However, these defect-related bSF, UVL, and BL peaks were observed only in the planar GaN layer sample and NP GaN sample without Ga<sub>2</sub>O<sub>3</sub>. We speculate that defect-related peaks are effectively suppressed by passivation with a thin Ga<sub>2</sub>O<sub>3</sub> layer surrounding NP GaN.

To clarify the passivation effect of the Ga<sub>2</sub>O<sub>3</sub> interlayer between the perovskite and NP GaN layers of the heterostructure, we spin-coated MAPbI<sub>3</sub> perovskite materials onto two NP GaN samples (with and without Ga<sub>2</sub>O<sub>3</sub>), and time-resolved PL (TR-PL) with steady-state PL measurements were carried out on both samples at room temperature (Figure 3e). The TR-PL spectrum of MAPbI<sub>3</sub> on NP GaN with Ga<sub>2</sub>O<sub>3</sub> showed a relatively short PL average lifetime (*t* = 10.07 ns), while MAPbI<sub>3</sub> on NP GaN without Ga<sub>2</sub>O<sub>3</sub> exhibited a longer PL lifetime (*t* = 18.20 ns, see Table S2). We attribute such short lifetime to the presence of Ga<sub>2</sub>O<sub>3</sub> between the MAPbI<sub>3</sub> and NP GaN layers, which facilitates the carrier extraction to the *n*-GaN. In the same vein, steady-state PL measurements indicated more pronounced PL quenching of MAPbI<sub>3</sub> on NP GaN with Ga<sub>2</sub>O<sub>3</sub> than on that without Ga<sub>2</sub>O<sub>3</sub>. Thus, introducing the Ga<sub>2</sub>O<sub>3</sub> interlayer is a useful strategy to suppress nonradiative recombination that occurs as a consequence of defect-related deep levels and energy-band mismatching, as shown in Figure 3f.

To further evaluate the effects of different porosities on NP GaN/MAPbI<sub>3</sub> PSCs, we compared the corresponding current–voltage characteristics of the samples using two solar cell characterization methods: *J*–*V* and external quantum efficiency (EQE) measurements. Figure 3g shows the current–voltage characteristics of our best-performing MAPbI<sub>3</sub> PSCs with planar *n*-GaN layer and NP GaN structures. The reverse-scan short-circuit current density (*J*<sub>sc</sub>), open-circuit voltage

(*V*<sub>oc</sub>), and fill factor (FF) of the solar cell containing planar *n*-GaN were 11.16 mA/cm<sup>2</sup>, 0.99 V, and 46.1%, respectively. The rigid PSCs containing a planar *n*-GaN layer exhibited a best PCE of 5.09%. In contrast, the MAPbI<sub>3</sub> PSCs containing NP GaN showed a considerable enhancement in *J*<sub>sc</sub>, *V*<sub>oc</sub>, and FF (see Table 1). The PCE of the solar cells increased

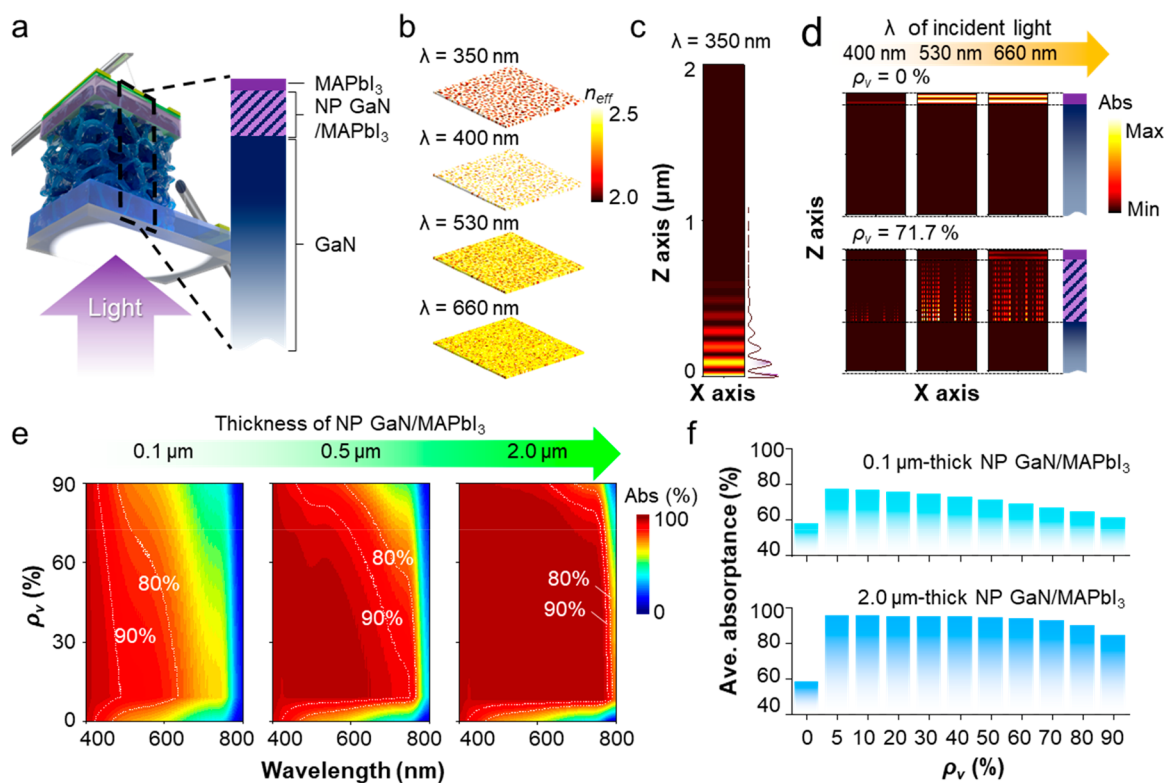
**Table 1. Summary of the *J*<sub>sc</sub>, *V*<sub>oc</sub>, FF, and PCE Parameters for PSCs with Planar GaN and NP GaN**

|                               | <i>J</i> <sub>sc</sub>   | <i>V</i> <sub>oc</sub> | FF    | PCE    |
|-------------------------------|--------------------------|------------------------|-------|--------|
| Planar GaN ( $\rho_s = 0\%$ ) | 11.16 mA/cm <sup>2</sup> | 0.99 V                 | 46.1% | 5.09%  |
| NP GaN ( $\rho_s = 7.2\%$ )   | 19.17 mA/cm <sup>2</sup> | 0.99 V                 | 67.2% | 12.75% |
| NP GaN ( $\rho_s = 47.5\%$ )  | 21.08 mA/cm <sup>2</sup> | 1.05 V                 | 72.8% | 16.12% |
| NP GaN ( $\rho_s = 71.7\%$ )  | 22.87 mA/cm <sup>2</sup> | 1.07 V                 | 76.8% | 18.79% |

continuously as  $\rho_s$  was increased from 7.2% to 47.5% and to 71.7%. When the  $\rho_s$  was increased to 71.7%, the rigid PSCs exhibited the best PCE of 18.79%. The corresponding EQE and integrated *J*<sub>sc</sub> of a representative solar cell containing NP GaN ( $\rho_s = 71.7\%$ ) is shown in Figure 3h. Figure S4 shows the detailed performance characteristics of our PSCs with NP GaN ( $\rho_s = 71.7\%$ ). To further understand the influence of the Ga<sub>2</sub>O<sub>3</sub> interlayer, we also fabricated solar cells from which the Ga<sub>2</sub>O<sub>3</sub> interlayer of the NP GaN structure was removed by chemical etching with BOE at room temperature for 5 min (Figure 3i). We observed that the presence of the Ga<sub>2</sub>O<sub>3</sub> interlayer between the NP GaN and perovskite layers, which formed spontaneously at the NP GaN surface during the electrochemical etching process, contributes to the increased solar cell efficiency. Our PSC with NP GaN/*n*-GaN exhibits the highest efficiency among published ITO-free PSCs with alternative TCLs (see Table S3).

To investigate the light absorption efficiency of the NP GaN/MAPbI<sub>3</sub>, we performed FDTD simulations. Figure 4a illustrates the simulated structure of the PSC with NP GaN/MAPbI<sub>3</sub> under UV and visible light illumination ( $\lambda = 350$ – $800$  nm). The dispersion and extinction coefficients of each material (GaN and MAPbI<sub>3</sub>) were taken from the reported values in the literature.<sup>59,60</sup> To model the effects of the nanopore distribution in the NP GaN, we designed the NP GaN in the FDTD simulations as repeating planes of porosity,  $\rho_p$ , such that the porosity per unit volume,  $\rho_v$ , is equivalent to its experimentally estimated value. Figure 4b shows the contrast difference in effective refractive index (*n*<sub>eff</sub>) of NP GaN/MAPbI<sub>3</sub> ( $\rho_s = 71.7\%$ ). The simulated *n*<sub>eff</sub> of NP GaN/MAPbI<sub>3</sub> shows an intermediate value between GaN's and MAPbI<sub>3</sub>'s intrinsic refractive indices at all incident lights ( $\lambda = 350$  nm –  $800$  nm), because the two homogeneously mixed materials follow the *n*<sub>eff</sub> based on the 2D Maxwell–Garnett theory<sup>61,62</sup> (see the Supporting Information for details). According to the FDTD simulations, Fresnel reflections are significantly suppressed in a PSC structure sequentially consisting of 2.0  $\mu$ m thick *n*-GaN, NP GaN/MAPbI<sub>3</sub>, and MAPbI<sub>3</sub> with a gradual refractive index change, compared to those of a PSC structure having an abrupt interface and, thus, decreasing photon losses.

Because UV light photocatalytically degrades the MAPbI<sub>3</sub><sup>63,64</sup>—and blocking it increases the lifespan of PSCs—we investigated the propagation of UV light ( $\lambda = 350$  nm) in the simulated PSC structure. Figure 4c shows that a 2.0  $\mu$ m thick *n*-GaN layer blocks incident UV light entirely



**Figure 4.** (a) Schematic diagram of the PSC modeled for the FDTD simulation. (b) Contrast difference in effective refractive index ( $n_{\text{eff}}$ ) of NP GaN/MAPbI<sub>3</sub> ( $\rho_s = 71.7\%$ ) according to the wavelength of incident light ( $\lambda = 350, 400, 530,$  and  $660$  nm). Simulated contour maps of the light absorption for (c) planar  $n$ -GaN layer with UV incident light ( $\lambda = 350$  nm) and (d) NP GaN/MAPbI<sub>3</sub> on  $n$ -GaN with white light ( $\lambda = 400, 530,$  and  $660$  nm).  $\rho_s = 0\%$  (top) consists of MAPbI<sub>3</sub> on  $n$ -GaN without NP GaN, and  $\rho_s = 71.7\%$  (bottom) consists of NP GaN/MAPbI<sub>3</sub> on  $n$ -GaN with a volume ratio of NP GaN (28.3%) and MAPbI<sub>3</sub> (71.7%) filling nanopores of the NP GaN. (e) Simulated contour maps for the absorption spectra of NP GaN/MAPbI<sub>3</sub> in the visible light region as functions of the porosity and thickness of NP GaN/MAPbI<sub>3</sub> light absorber. Two dashed lines in contour maps indicate the criteria for 80% and 90% of the incident light absorbance. (f) The averaged light absorbance of 0.1 (top) and 2.0  $\mu\text{m}$ -thick (bottom) NP GaN/MAPbI<sub>3</sub> with varying  $\rho_v$ , respectively.

from reaching the NP GaN/MAPbI<sub>3</sub> or MAPbI<sub>3</sub> perovskite light absorber layers.

In contrast, light within the visible spectrum ( $\lambda = 400, 530,$  and  $660$  nm) penetrated the PSC structures and was only absorbed by the perovskite within the MAPbI<sub>3</sub> and NP GaN/MAPbI<sub>3</sub> layers (see Figure 4d for the simulated absorption profiles). Interestingly, the PSC with NP GaN/MAPbI<sub>3</sub> shows an extended absorption distribution along the entire NP GaN/MAPbI<sub>3</sub>, implying that the hierarchical configuration of NP GaN/MAPbI<sub>3</sub> further increases the probability of light absorption.

To analyze the porosity- and thickness-dependent light absorption efficiencies, we performed RCWA simulations. Figure 4e shows the contour maps of calculated light absorbance ( $\lambda = 400\text{--}800$  nm) by increasing the thickness of NP GaN/MAPbI<sub>3</sub> and the porosity of NP GaN. The two dashed white lines demarcate the region of the graph for 80% and 90% of the incident light absorbance according to the porosity of NP GaN. This result reveals that when the porosity of NP GaN is between 10% and 80%, the structure shows high absorption for visible light. Moreover, when the NP GaN/MAPbI<sub>3</sub> light absorber has a thickness of more than 2.0  $\mu\text{m}$ , perfect absorption occurs throughout the visible light regions. Figure 4f shows the average light absorbance of 0.1 and 2.0  $\mu\text{m}$ -thick NP GaN/MAPbI<sub>3</sub> across the entire visible wavelengths. The simulation results suggest that the optimal porosity of NP GaN and the optimal thickness of the NP

GaN/MAPbI<sub>3</sub> should be between 10% and 80% and  $\geq 2.0$   $\mu\text{m}$ , respectively.

In conclusion, the cathode structure in a PSC plays a key role in the extraction of photogenerated electrons. Therefore, one of the most critical aspects of PSC fabrication is finding a suitable combination of a TCL and an ETL, which are typically composed of differing materials, to form the cathode structure. In this respect, we proposed a novel cathode structure for PSCs: a NP GaN/ $n$ -GaN that combines the function of both the TCL and ETL. The NP GaN/ $n$ -GaN structure affords enhanced light harvesting (as a result of the structure's gradual refractive index change) and efficient carrier extraction due to the enlarged perovskite interface with the cathode. A Ga<sub>2</sub>O<sub>3</sub> interlayer (formed spontaneously at the surface of the NP GaN layer during the electrochemical etching process) between NP GaN and perovskite contributes to interface passivation. These advantageous properties enabled the fabrication of MAPbI<sub>3</sub> PSCs based on NP GaN/ $n$ -GaN structures that show promising PCEs of up to 18.79%. Our work opens an avenue for PSCs to benefit from the versatile heterostructures afforded by the mature III-nitride-based technologies.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsenerylett.0c01621>.

Experimental details of *n*-GaN growth, electrochemical etching process, TEM, device fabrication, current–voltage characteristics, EQE measurement, FDTD simulation, PL, and TRPL (PDF)

## AUTHOR INFORMATION

### Corresponding Authors

**Boon S. Ooi** – Photonics Laboratory, Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; Email: boon.ooi@kaust.edu.sa

**Osman M. Bakr** – Division of Physical Sciences and Engineering and KAUST Catalyst Center (KCC), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0002-3428-1002](https://orcid.org/0000-0002-3428-1002); Email: osman.bakr@kaust.edu.sa

### Authors

**Kwang Jae Lee** – Division of Physical Sciences and Engineering and KAUST Catalyst Center (KCC), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0002-3158-7532](https://orcid.org/0000-0002-3158-7532)

**Jung-Wook Min** – Photonics Laboratory, Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0002-7570-1739](https://orcid.org/0000-0002-7570-1739)

**Bekir Turedi** – Division of Physical Sciences and Engineering and KAUST Catalyst Center (KCC), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

**Abdullah Y. Alsalloum** – Division of Physical Sciences and Engineering and KAUST Catalyst Center (KCC), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0001-8988-1307](https://orcid.org/0000-0001-8988-1307)

**Jung-Hong Min** – Photonics Laboratory, Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

**Yeong Jae Kim** – School of Electrical Engineering and Computer Science, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea; [orcid.org/0000-0002-0870-9044](https://orcid.org/0000-0002-0870-9044)

**Young Jin Yoo** – School of Electrical Engineering and Computer Science, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea

**Semi Oh** – School of Materials Science and Engineering, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea

**Namchul Cho** – Department of Energy Systems Engineering, Soonchunhyang University, Asan 31538, Republic of Korea

**Ram Chandra Subedi** – Photonics Laboratory, Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0003-2818-1352](https://orcid.org/0000-0003-2818-1352)

**Somak Mitra** – Semiconductor and Material Spectroscopy Laboratory, Material Science & Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

**Sang Eun Yoon** – Department of Molecular Science and Technology, Ajou University, Suwon 16499, Republic of Korea  
**Jong H. Kim** – Department of Molecular Science and Technology, Ajou University, Suwon 16499, Republic of Korea; [orcid.org/0000-0001-6782-2154](https://orcid.org/0000-0001-6782-2154)

**Kwangwook Park** – Division of Advanced Materials Engineering, Jeonbuk National University, Jeonju 54896, Republic of Korea; [orcid.org/0000-0002-4600-4600](https://orcid.org/0000-0002-4600-4600)

**Tae-Hoon Chung** – Light Source Research Division, Korea Photonics Technology Institute (KOPTI), Gwangju 61007, Republic of Korea

**Sung Hoon Jung** – Light Source Research Division, Korea Photonics Technology Institute (KOPTI), Gwangju 61007, Republic of Korea

**Jong H. Baek** – Light Source Research Division, Korea Photonics Technology Institute (KOPTI), Gwangju 61007, Republic of Korea

**Young Min Song** – School of Electrical Engineering and Computer Science, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea; [orcid.org/0000-0002-4473-6883](https://orcid.org/0000-0002-4473-6883)

**Iman S. Roqan** – Semiconductor and Material Spectroscopy Laboratory, Material Science & Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0001-7442-4330](https://orcid.org/0000-0001-7442-4330)

**Tien Khee Ng** – Photonics Laboratory, Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia; [orcid.org/0000-0002-1480-6975](https://orcid.org/0000-0002-1480-6975)

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsenerylett.0c01621>

### Author Contributions

<sup>†</sup>K.J.L. and J.-W.M. contributed equally to this work.

### Notes

The authors declare no competing financial interest.

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