Supplementary Information

Plasma-Enhanced Atomic-Layer-Deposited Gallium Nitride as an Electron Transport Layer for Planar Perovskite Solar Cells

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Fig.S1 HRTEM image of GaN thin-film on Si(100) substrate.



Fig. S2 Plots of $(\alpha hv)^2$ versus hv for the absorption spectra.



Fig.S3 XPS spectra from the 200-PEALD-cycle-GaN thin film.



Fig. S4 Contact angle measurement with *N*, *N*-dimethylformamide (DMF) and dimethylsulfoxide (DMSO) solutions (perovskite solvents) on GaN films.



Fig. S5 (a) SEM images of the perovskite films grown on different substrates: (a) bare-FTO glass; (b) 10-PEALD-cycle GaN; (c) 30-PEALD-cycle GaN; (d) 50-PEALD-cycle GaN; (e) 80-PEALD-cycle GaN (The difference between above SEM images and Fig. 5c arises from to the change of some chemical reagents in our lab.).



Fig. S6 (a) Top-view SEM image of the perovskite film deposited on TiO_2 ETL surface; (b) *J-V* curve of the TiO_2 ETL-based PSC.



Fig. S7 Statistic histogram of the efficiency of the devices without and with 50-PEALD-cycle GaN layer.



Fig. S8 Long-term stability tests for the GaN-free and 50-PEALD-cycle PSCs. Both the devices were constantly illuminated under maximum power point (65°C, ~30 RH%) in nitrogen atmosphere without encapsulation.

The PL decay can be well fitted with the double-exponential function, and the average emission lifetime is determined by the following equation:

$$\tau_{\text{ave}} = \left(A_1 \tau_1^2 + A_2 \tau_2^2 \right) / \left(A_1 \tau_1 + A_2 \tau_2 \right)$$

where τ_1 and τ_2 are the lifetimes, A_1 and A_2 are their amplitudes, and τ_{ave} is the average lifetime. The corresponding fitting results are summarized in Table S1.

 Table S1 Fitting results of the time-resolved PL decay curves with bi-exponential functions

Samples	A_1	τ_1 (ns)	A_2	τ_2 (ns)	$\tau_{ave}(\mathrm{ns})$
GaN-free	0.954	22.04	0.146	88.21	47.17
50-PEALD-cycle GaN	1.172	4.95	0.280	32.07	21.42

Samples	$R_{s}\left(\Omega ight)$	$R_{ct}(\mathbf{k}\Omega)$	$R_{rec}(\mathbf{k}\Omega)$
GaN-free	22.1	87.5	137.5
50-PEALD-cycle GaN	22.7	82.6	169.1

Table S2 EIS fitting results derived from the Nyquist plots of the PSCs