Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Gallium Nitride (GaN) Protected Tandem Photoelectrodes for High Efficiency, Low Cost, and Stable Solar Water Splitting

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DOE Project Award # (DE-EE0010730)

Date: May 8, 2023

Project ID: P209

DOE Hydrogen Program 2024 Annual Merit Review and Peer Evaluation Meeting

NREL Transforming ENERGY | BERKELEY LAB Bringing Science Solutions to the World | Sandia National Laboratories | INL Idaho National Laboratory | Lawrence Livermore National Laboratory

Project Overview

Project Partners

Zetian Mi, University of Michigan, Ann Arbor
Yanfa Yan, University of Toledo
Pallab Bhattacharya, University of Michigan, Ann Arbor
James Young, Todd Deutsch, Myles Steiner, and Kai Zhu, National Renewable Energy Laboratory
Tadashi Ogitsu, Lawrence Livermore National Laboratory
Joel Aber, Lawrence Berkeley National Laboratory

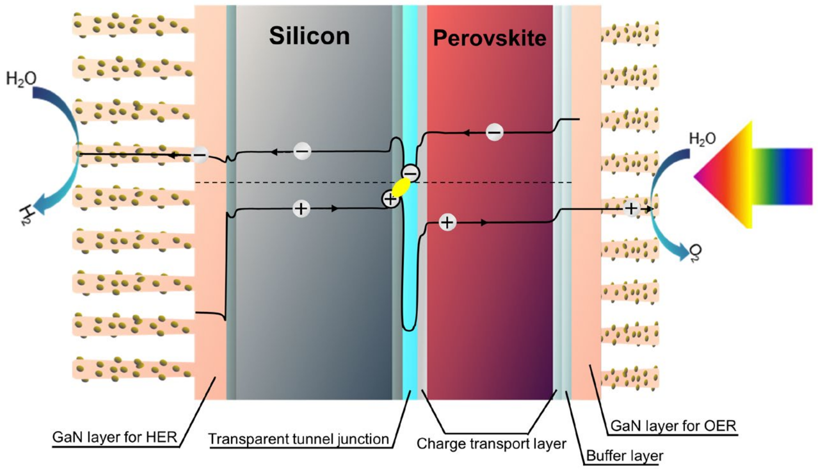
Award #	EE0010730
Start/End Date	10/01/2023 – 09/31/2026
Project Funding*	$1.2M

Project Vision

We propose to develop GaN-protected multi-junction tandem photoelectrodes to achieve both high efficiency (STH >20%) and long-term stable (1,000 h) spontaneous water splitting systems.

Project Impact

The success of the project will help meet the DOE 2026 target and pave the way for widespread commercialization of solar hydrogen production technologies.



A wireless Si/perovskite photoelectrode protected by GaN.

** this amount does not cover support for HydroGEN resources leveraged by the project (which is provided separately by DOE)*

Approach – Summary

Project Motivation

- To date, no PEC water splitting devices can simultaneously achieve both high efficiency (>10%) and stable operation (>200 h).
- We have previously demonstrated GaN-based photoelectrodes can exhibit long-term stability (3,000 h), due to the formation of a GaON protection layer.
- We will integrate GaN with high efficiency perovskite and III-V photoelectrodes to achieve stable operation.

Key Impact

Metric	State of the Art	Expected Advance
STH Efficiency	*10-20%*	*15-20%*
Stability	*<200 h*	*>1,000 h*

Barriers

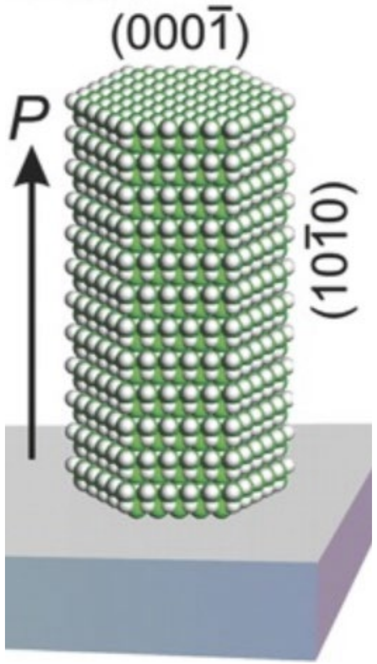
- Processing compatibility. High efficiency perovskites and III-V materials are generally synthesized at low temperatures (<500 ˚C), whereas GaN is grown at high temperature (>700 ˚C). The major barrier is how to achieve low temperature deposition of ultrathin GaN without compromising the surface protection and efficiency.

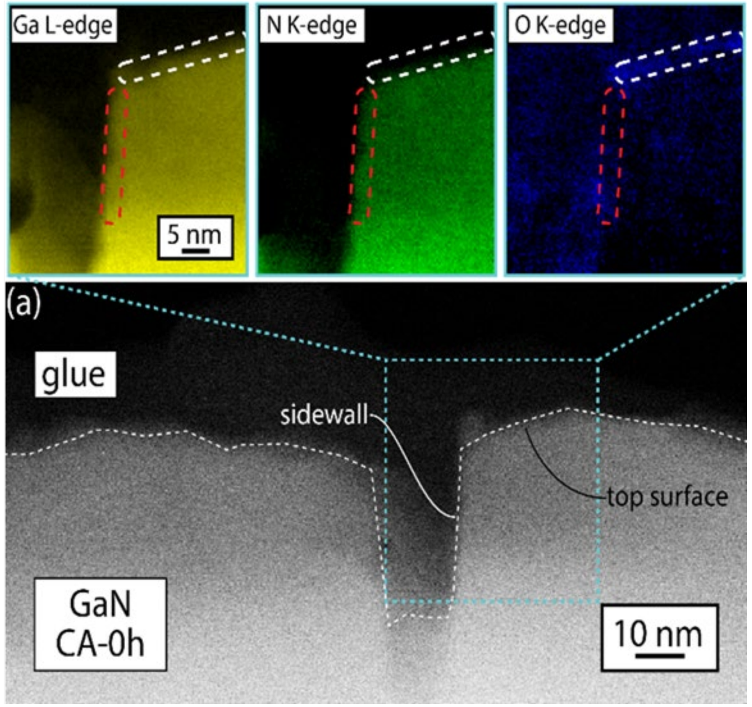
Partnerships

- K. Zhu and J. Berry, NREL; Synthesis of perovskites
- M. Steiner, NREL; Epitaxial growth of III-V wafers
- James Young and T. Deutsch, NREL; PEC characterization and testing
- T. Ogitsu, LLNL; Electrochemical interface studies
- J. Ager, LBNL; Structural and interface characterization
- Y. Yan, Toledo; Synthesis of perovskites and PEC testing

Approach – Innovation

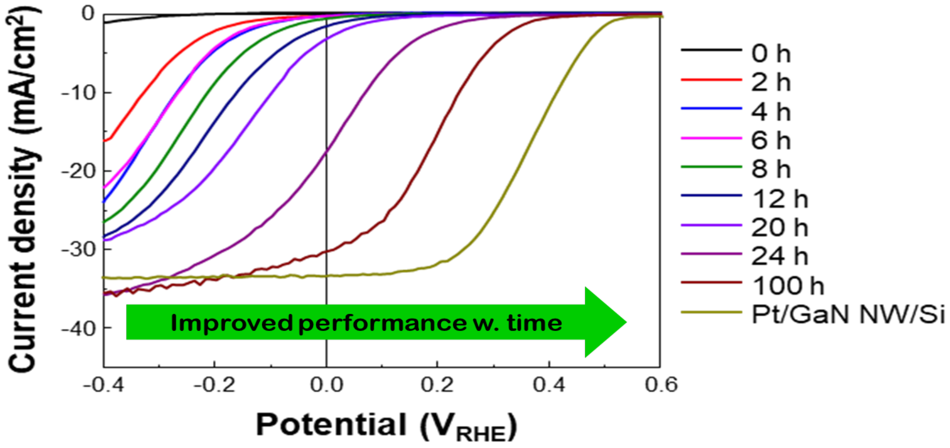
- **We have discovered that the non-polar surfaces of GaN can be transformed to a stable, atomically thin GaON layer during harsh PEC reaction.**

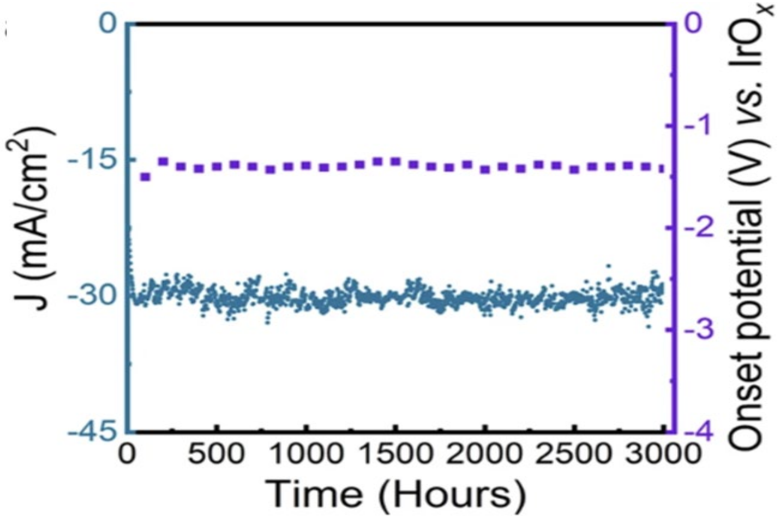




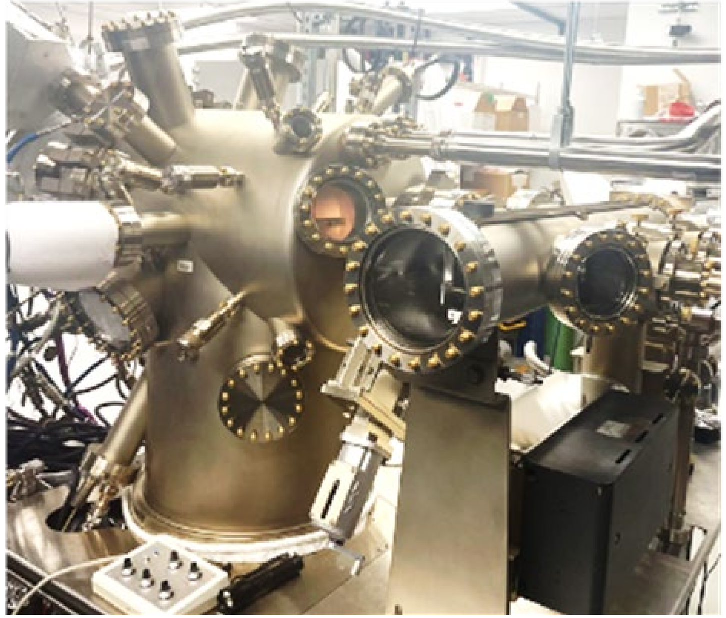
Nature Materials 20, 1130 (2021)
Nature Communications 14, 2047 (2023)

- **The formation of GaON leads to self-improved performance and long-term stability (3,000 h).**



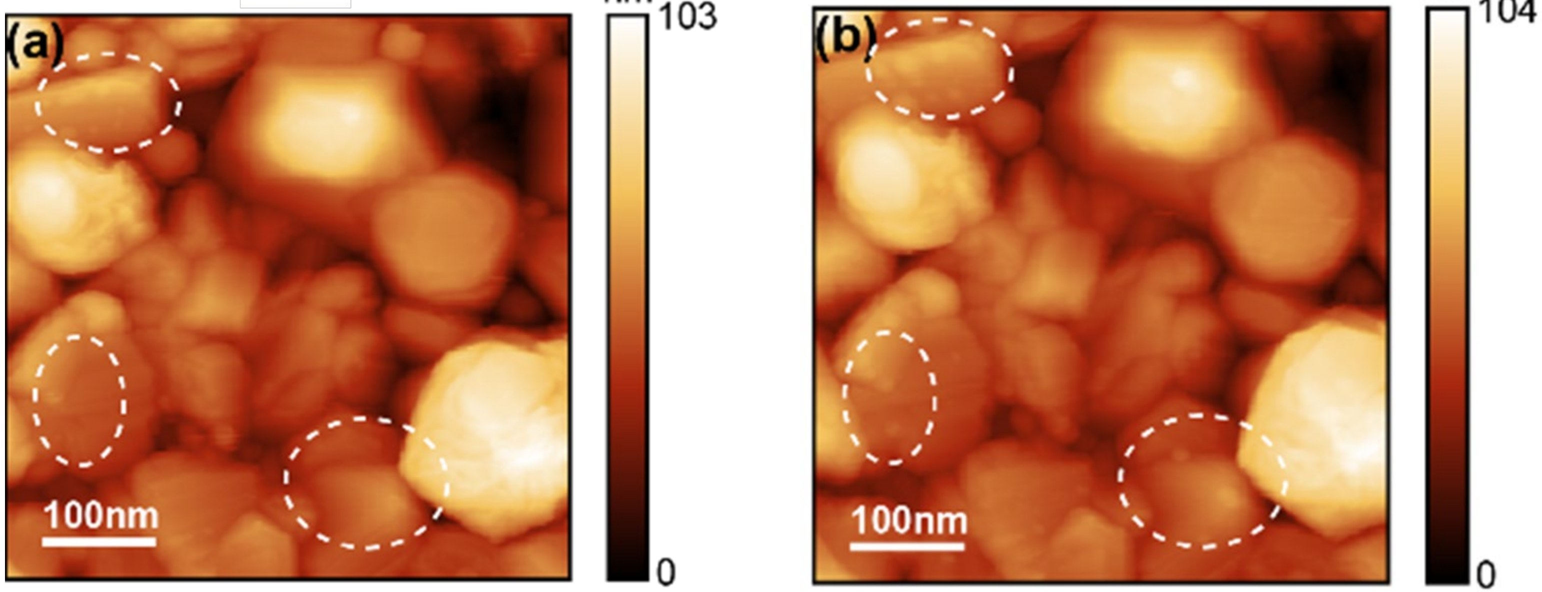


- **To address the processing incompatibility between III-V (or perovskites) with GaN, we utilize a low-temperature epitaxial growth process and nanoscale engineering to achieve high quality GaN surface protection that was not possibly previously.**



Molecular beam epitaxy

0 hr *120 mins*



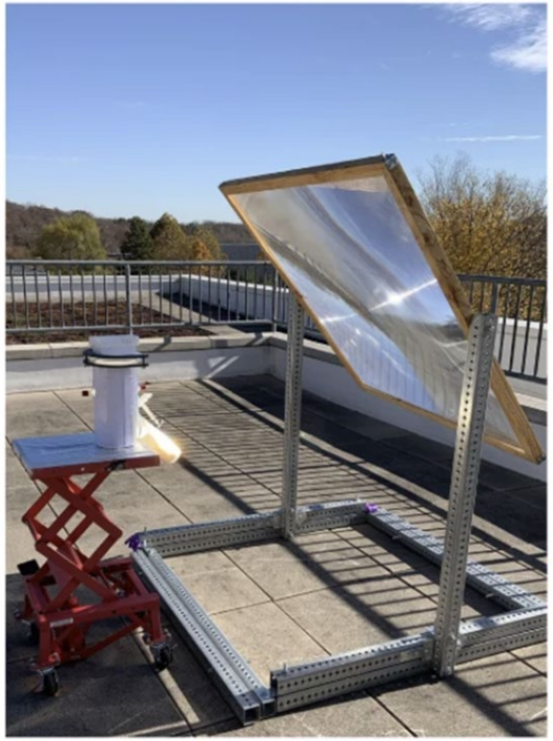
No degradation was observed for GaN nanostructures during water splitting

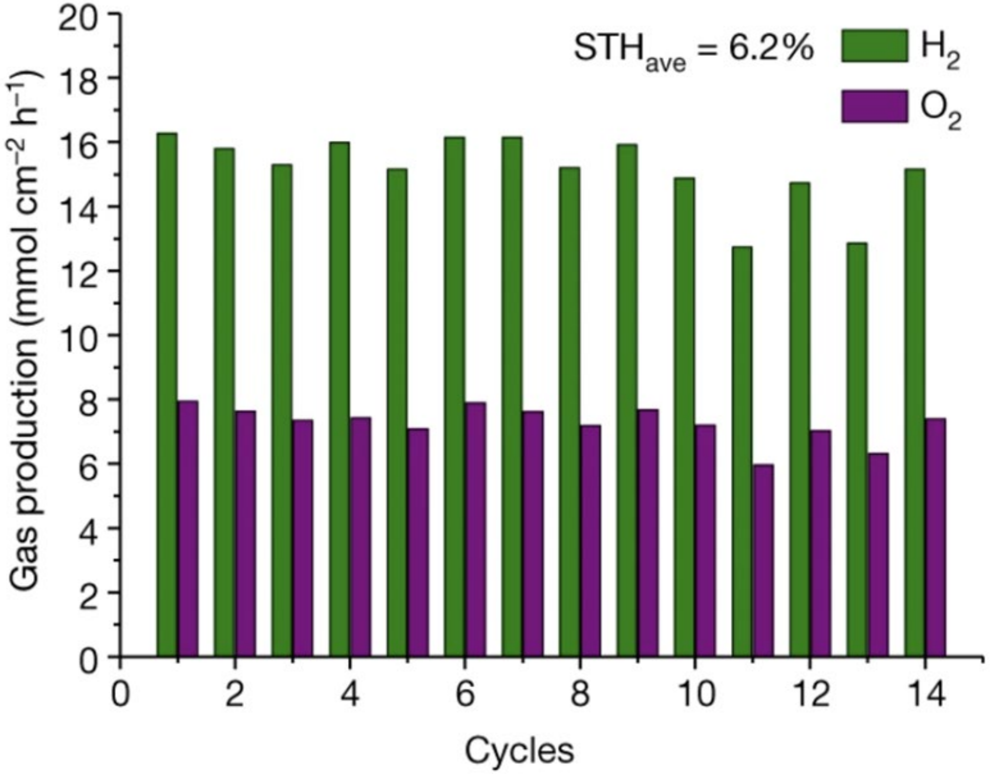
Nature Materials 20, 1130–1135 (2021); Nature Communications 14, 2047 (2023)

Approach – Innovation

- **Concentrated sunlight will be utilized to significantly reduce the amount of photocatalyst and therefore the overall system cost.**

Nature 613, 66-70, 2023.





With the use of GaN-based nanostructures, we have recently demonstrated direct solar water splitting with STH of 6.2% under 160x concentrated sunlight.

Approach – Safety Planning and Culture

Safety plan was required

1. Safety plan was submitted to be reviewed by HSP

Prioritizes safety and analyzes safety hazards

1. The PI will lead the development of this safety analysis and ensuing Risk Assessment/Ranking/Plan.
2. EHS expertise leads the project team through the processes of identifying, categorizing, and ranking hazards and developing mitigation solutions.
3. All hazard analysis documentation will be approved by the PI, Project Lead, and EHS manager.

Addresses incidents and near-misses; and/or

1. Monthly safety meeting address incidents, near-misses and issues affecting safety changes.

Incorporates best safety practices and lessons learned.

1. All personnel who work with these systems are trained and have working knowledge of all hazards that pertain to a project prior to beginning work.
2. New and refresher safety training needs are identified by the Project Leader and the EHS Department.
3. Training records are maintained by the EHS Department and are available to the Project Leader.

Potential Impact

- The STH conversion efficiency and cost targets for purified, 300 psi compressed H_2 gas are 20% STH and \$1 per kg H_2 by 2030. The tandem PEC device concept of stacking wide-bandgap and narrow-bandgap semiconductors is a proven method that can achieve the targeted STH efficiency. To date, however, the major limiting factor is the poor stability for high efficiency devices. We have demonstrated that GaN nanostructures can protect against photocorrosion and oxidation without affecting PEC performance. If successful, this project will enable the simultaneous achievement of PEC water splitting devices with both high efficiency and long-term stability.
- The use of low cost, high efficiency, and scalable perovskite light absorber can significantly reduce the cost for PEC water splitting systems.
- The cost issue can be further reduced by utilizing concentrated sunlight, which can significantly reduce the amount of semiconductor light absorber and catalyst materials used in solar water splitting systems. With the use of GaN nanostructures, we have recently demonstrated direct solar water splitting with an STH efficiency of 6.2% under concentrated (160x) sunlight illumination.
- The outcome of this project is to develop GaN-protected tandem photoelectrodes, with the objective to achieve high efficiency (up to 20%) and long-term stability (>1,000 hours) solar-to-H_2 conversion through PEC water splitting. The success of this project will help meet the DOE technical target for H_2 production from PEC water splitting.

Potential Impact

This project leverages the existing unique expertise and capabilities of HydroGEN Energy Materials Network (EMN). We have been working closely with the following to advance the proposed project:

1) Probing and Mitigating Chemical and Photochemical Corrosion of Electrochemical and Photoelectrochemical Assemblies, Joel Ager, LBNL. With the unique *in situ* techniques, including photoelectrochemical AFM and STM, our previously collaboration with LBNL revealed that GaN/Si photoelectrodes, without any extra surface protection, showed no sign of any performance degradation during continuous PEC water splitting, which is in stark contrast for the rapid degradation of conventional high efficiency photoelectrodes. The underlying mechanisms have also been identified.
2) Ab Initio Modeling of Electrochemical Interfaces, Tadashi Ogitsu, LLNL. This collaboration provides important insights of electrochemical interface and PEC device optimization through ab-initio modeling and computational materials diagnostics. Preliminary studies revealed the underlying mechanism for the extraordinary stability of N-rich GaN-based photoelectrodes.
3) Scaled up PEC testing, and surface Modifications for Catalysis and Corrosion Mitigation, James Young and Todd Deutsch, NREL. Dr. Deutsch and his team members have been working on the co-catalyst deposition and surface protection to identify the best strategy to protect the surface against photocorrosion and oxidation. Moreover, Dr. Deutsch will work with the team for PEC performance characterization, validation, and scaled up testing.
4) Perovskite materials, Kai Zhu, NREL. Dr. Zhu and his team members have been working with us by providing high efficiency perovskite materials and tandem cells for integration with GaN surface protection scheme and PEC testing.
5) III-V tandem devices, Myles Steiner, NREL. Dr. Steiner and his team members have been working with us by providing high efficiency III-V tandem devices for integration with GaN surface protection scheme and PEC testing.

Accomplishments

Budget Period 1 Scope of Work and Go/No Go Milestones

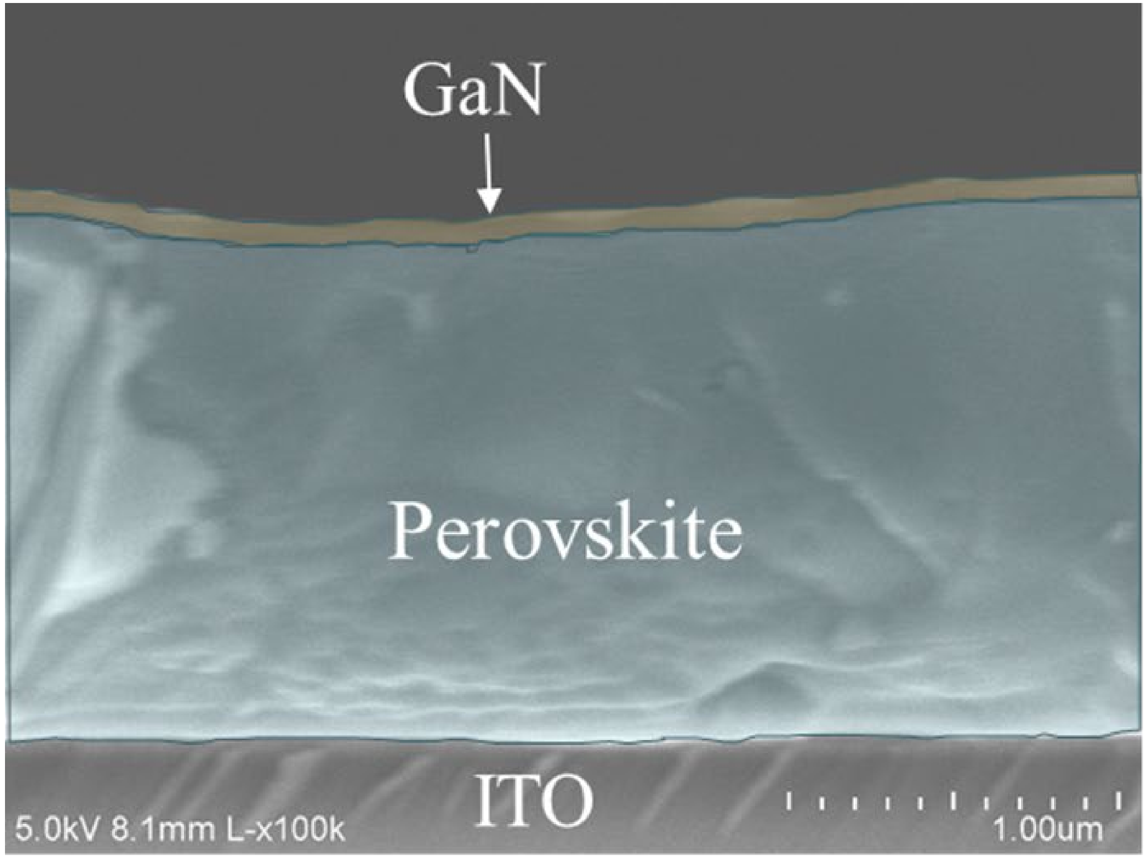
- We will demonstrate GaN-protected perovskite/Si tandem device and GaN-protected GaInP/GaInAs tandem photoelectrodes. A wide range of synthesis techniques, including spin-coating, sputtering, and MBE will be utilized.
- <u>We will not proceed if we cannot achieve:</u> GaN-protected tandem device with photocurrent density of >12.5 mA/cm^2, OCP of >1.5 V, and stability of maintaining 80% the initial current for >100 h.

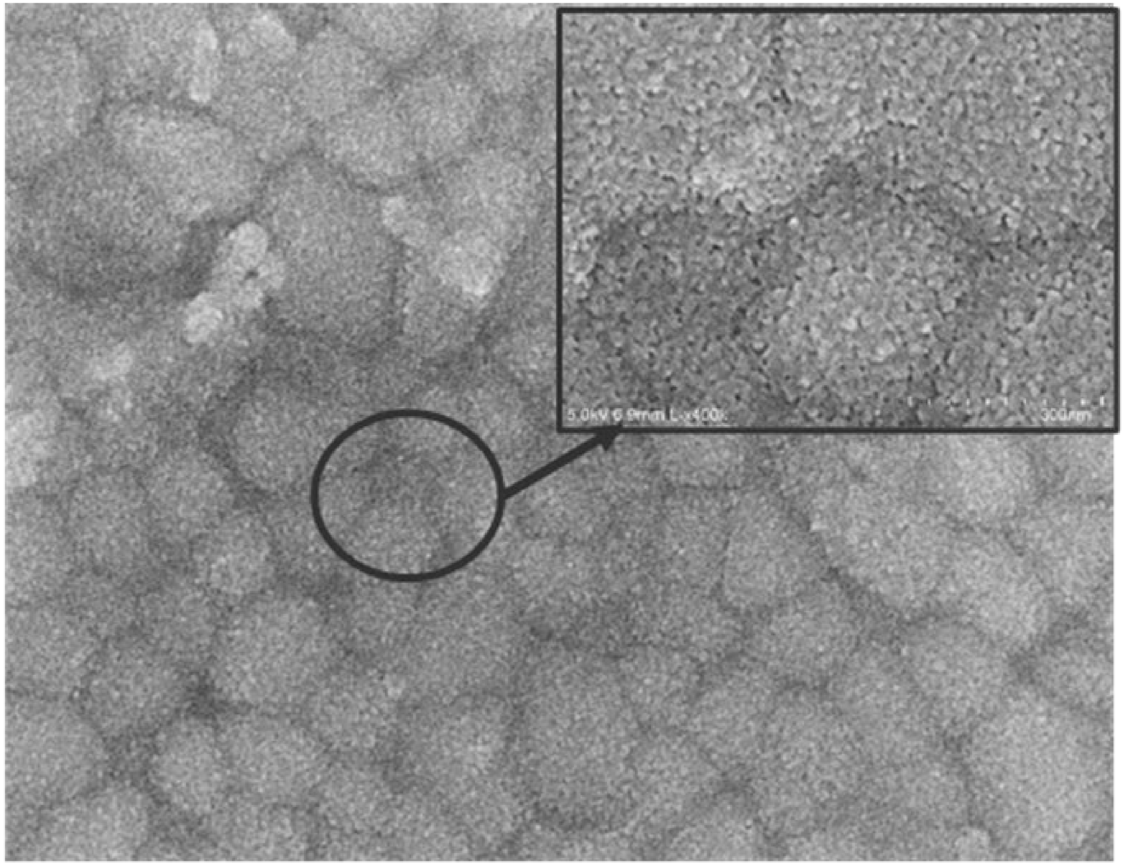
Significance of Meeting the Milestones

- The realization of GaN-protected tandem device with high performance will verify our proposed concept of utilizing GaN nanostructures to simultaneously achieve high efficiency and stable solar water splitting that was not previously possible.
- Collaborations with K. Zhu, M. Steiner, and T. Deutsch at NREL, J. Ager at LBNL, and T. Ogitsu and LLNL are established, enabling effective materials exchange, integration, characterization, and testing needed for theoretical and experimental understanding of the device design and performance.
- Meeting the budget period 1 Go/No Go milestones will help us to achieve unassisted solar water splitting with >20% STH efficiency and stability >1,000 h for future low-cost H_2 production.

Accomplishments

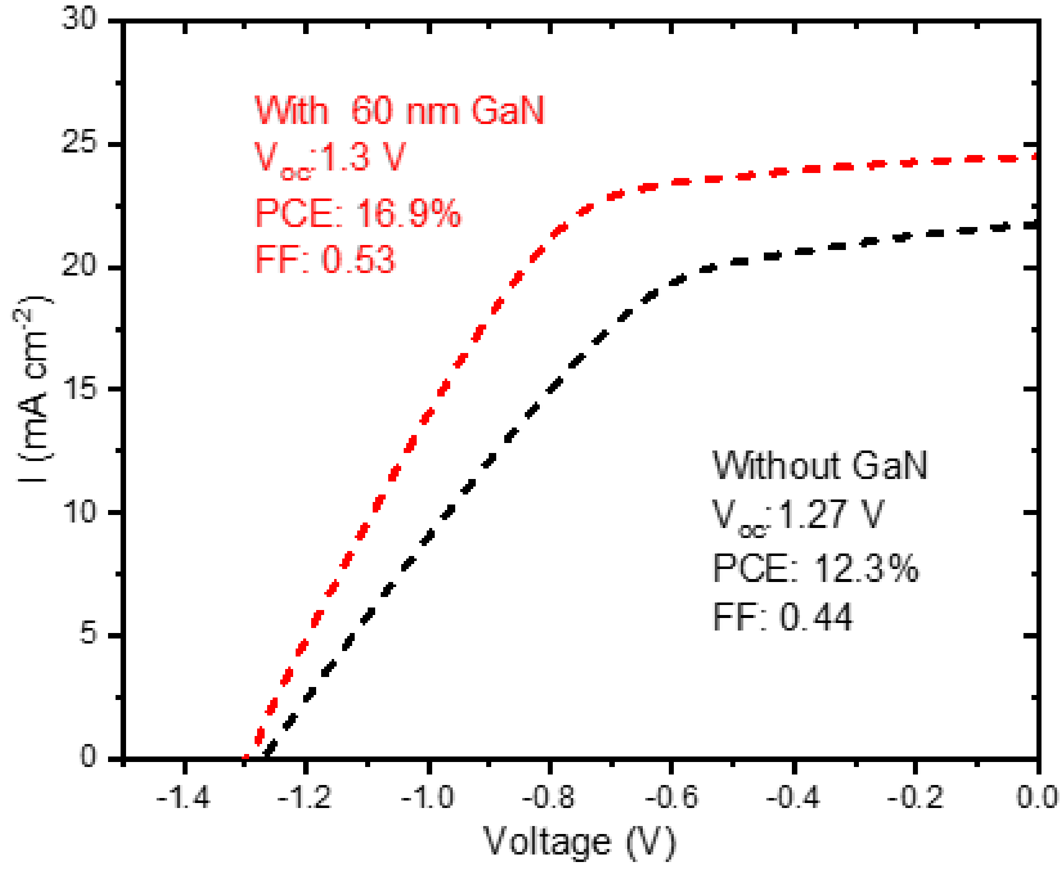
- **Successful demonstration of uniform GaN coverage on perovskite utilizing a low-temperature growth process**

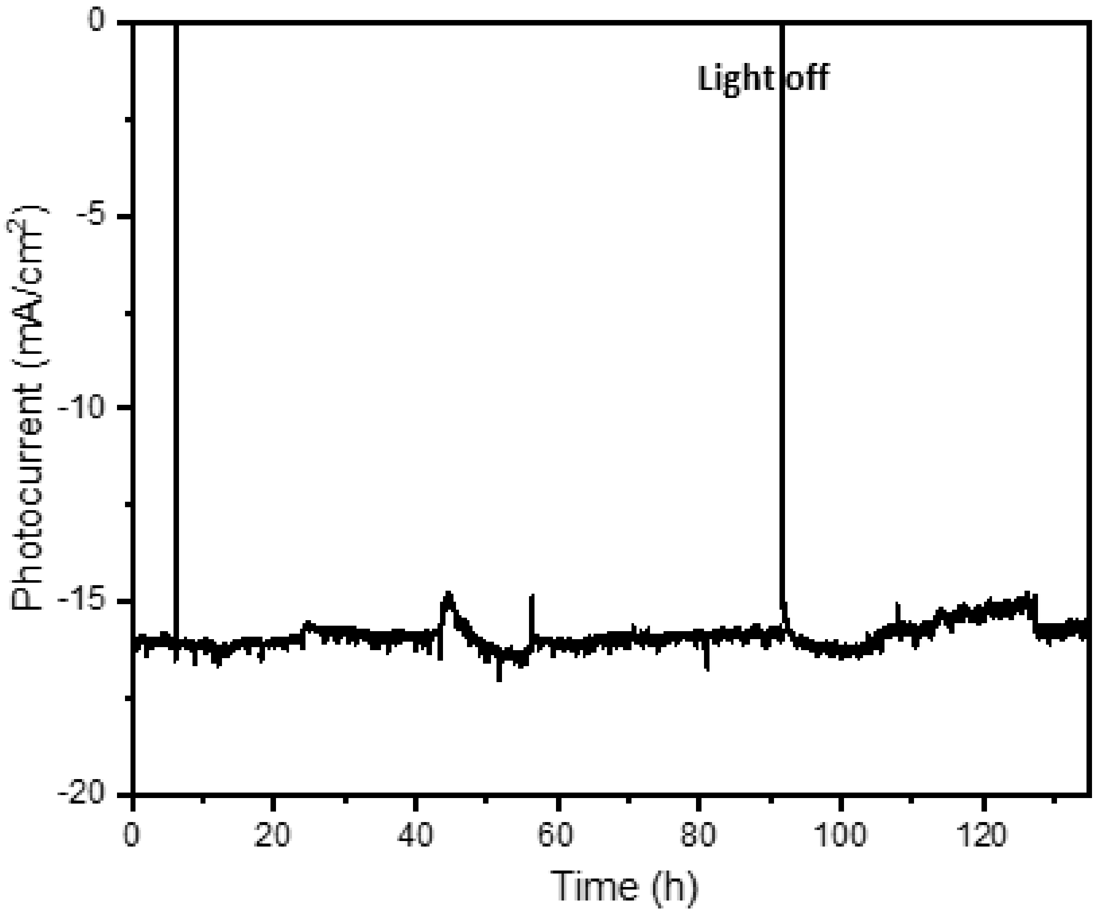




Accomplishments

➢ Efficient and stable GaN-protected perovskite device

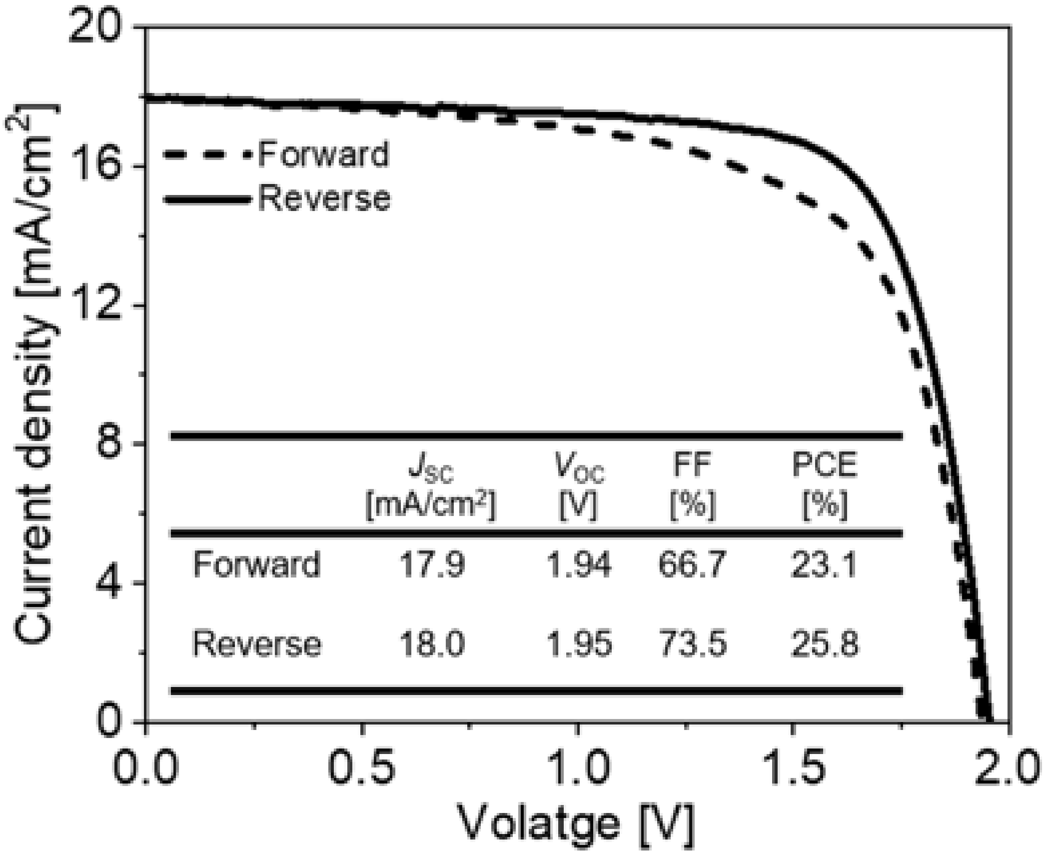




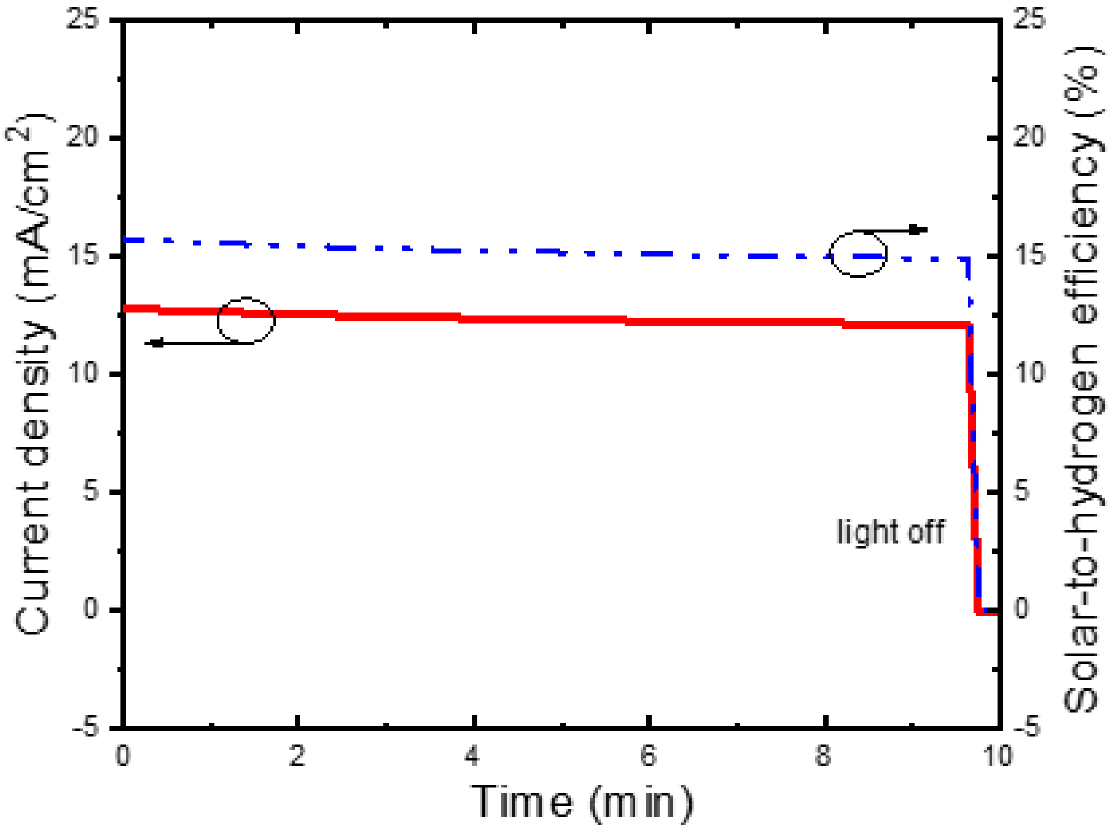
Encapsuled GaN-protected perovskite solar cell enables 100 h water splitting.

Accomplishments

➢ High performance perovskite/Si tandem device



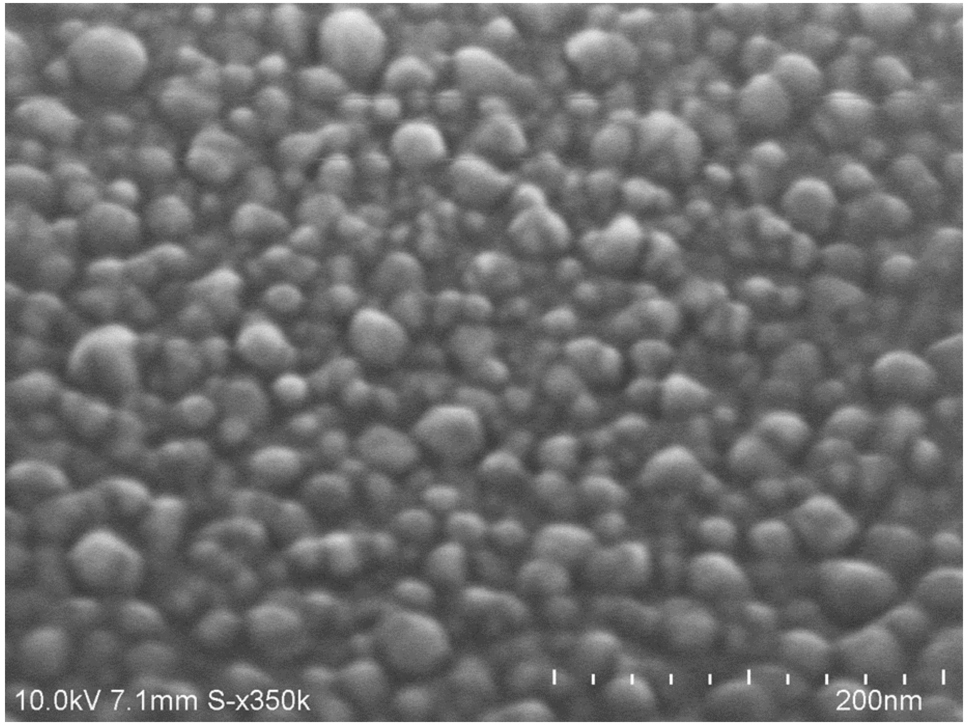
	J_{SC} [mA/cm²]	V_{OC} [V]	FF [%]	PCE [%]
Forward	17.9	1.94	66.7	23.1
Reverse	18.0	1.95	73.5	25.8

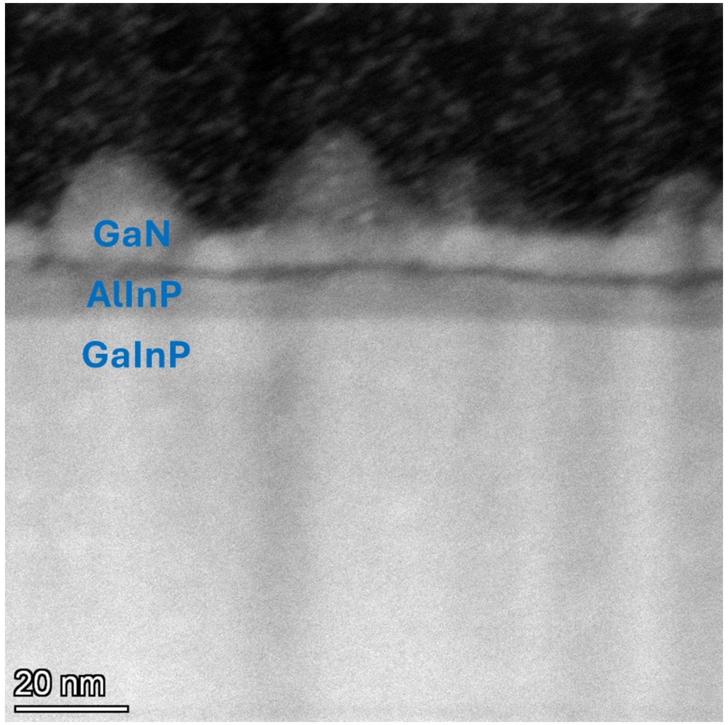


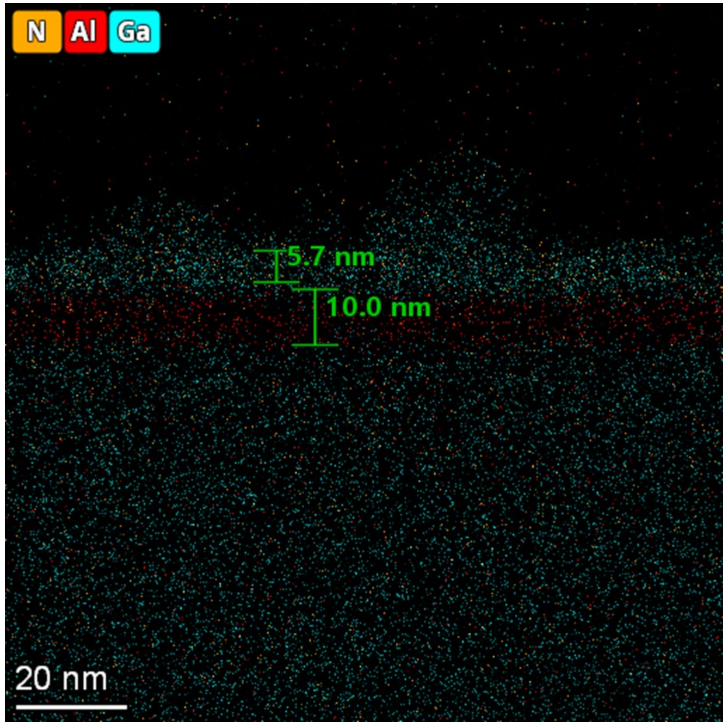
Initial studies showed perovskite/Si tandem devices exhibited a V_{oc} of 1.9 V, J_{sc} > 17 mA/cm², leading to a STH efficiency of 15.6%.

Accomplishments

- Successful deposition of GaN surface protection layer on GaInP/InGaAs photoelectrode



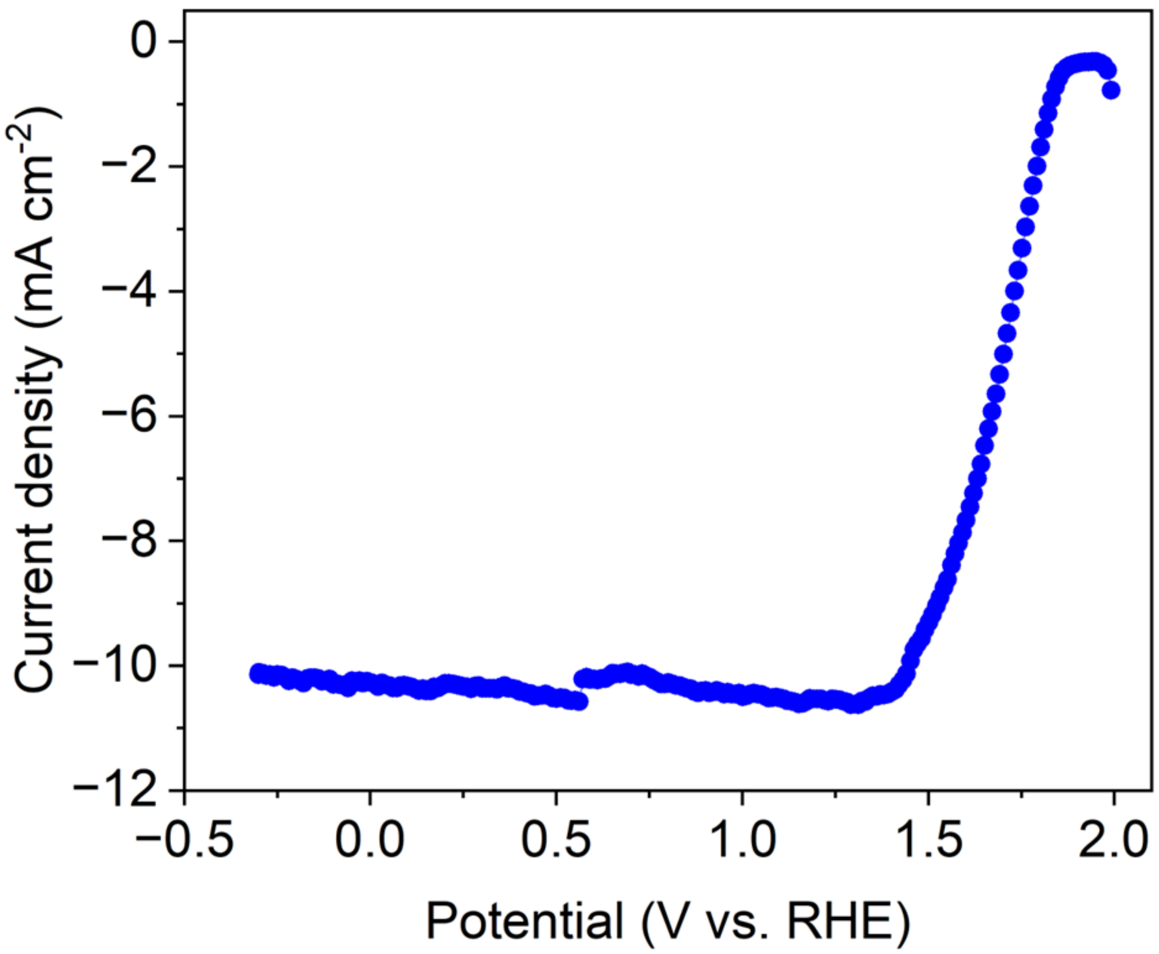


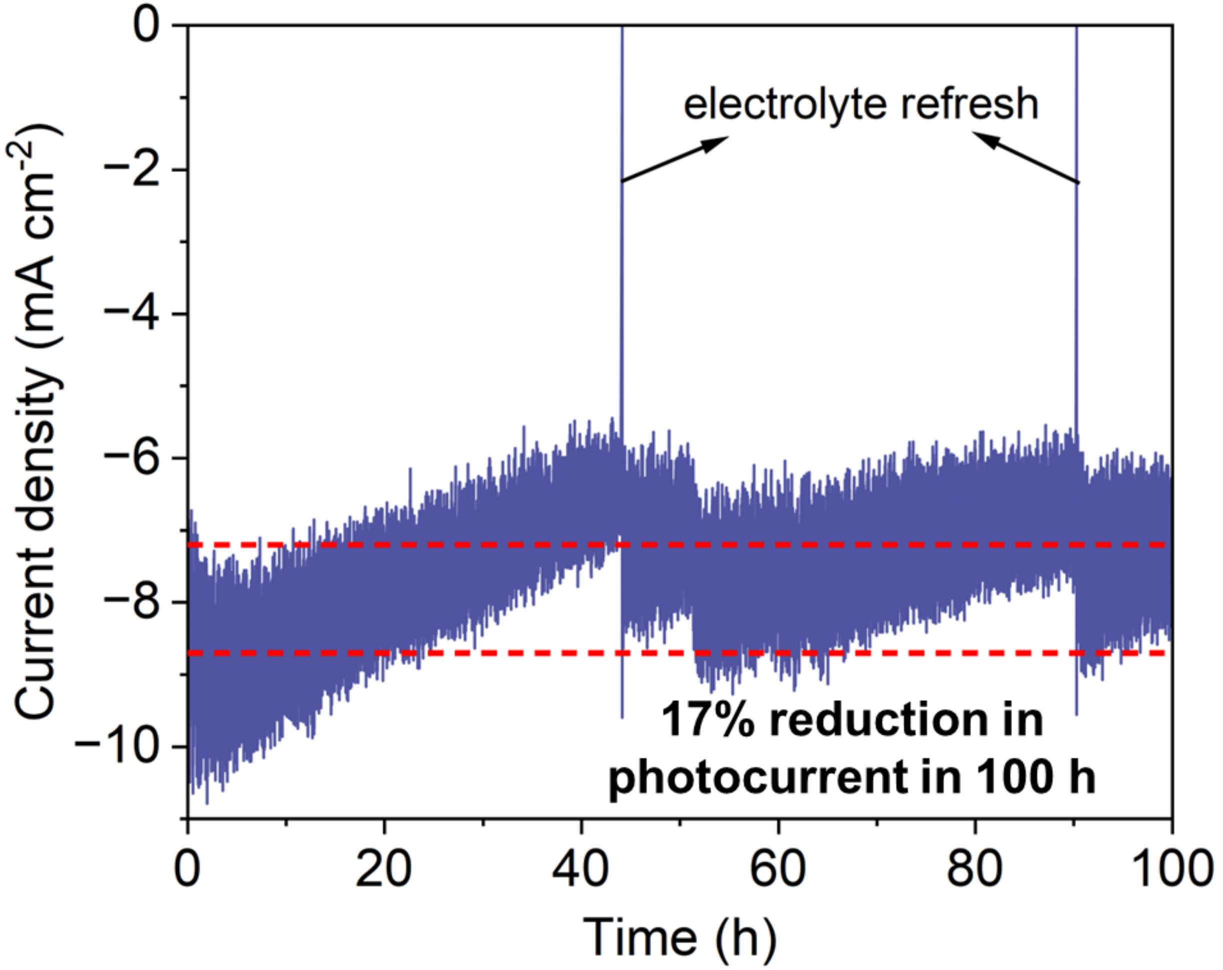


GaN surface protection layer with uniform coverage was grown on GaInP/GaInAs photoelectrode by MBE. The surface morphology and properties of GaN can be further optimized by varying the growth conditions.

Accomplishments

- **GaN protected GaInP/InGaAs photoelectrode showed stable operation with T80 >100 h**





Accomplishments

Outlook and Projected Outcomes

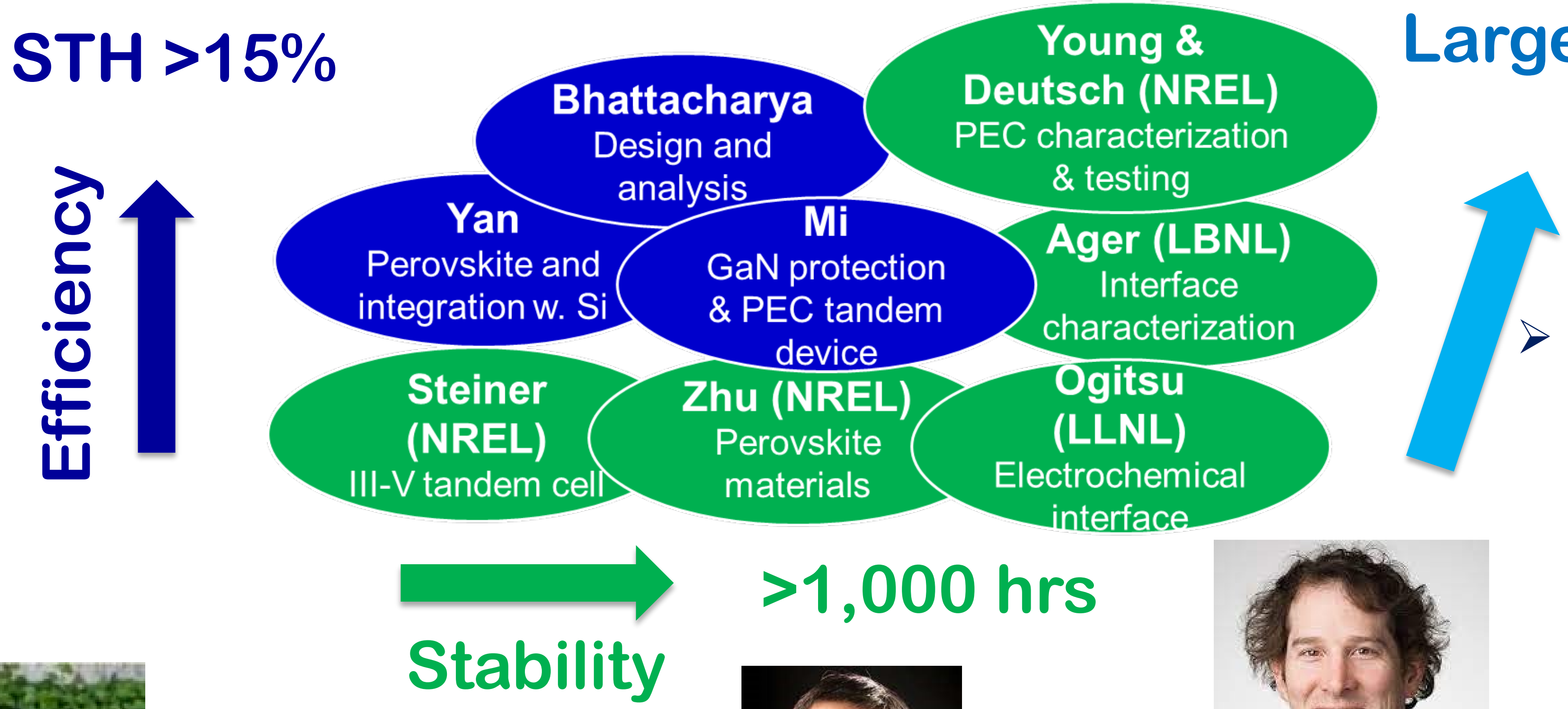
- Optimize the deposition conditions of GaN nanostructures on perovskite and III-V tandem devices to minimize the formation of pinholes and cracks to achieve STH efficiency >15% and long-term stable operation in two-electrode configuration.
- To further evaluate the performance under concentrated sunlight illumination conditions.

Confidence in meeting the Go/ No Go milestones: High

Major Impact

- This project will be instrumental to establish a low-cost platform for high efficiency PEC water splitting systems for green hydrogen production.
- The stability of PEC water splitting devices will be fundamentally improved by utilizing N-terminated GaN protection layer.
- The semiconductor photoelectrodes are synthesized using industry ready materials, e.g., perovskite, III-V, Si and GaN based, and therefore the manufacture is controllable, scalable, and of low cost.

Collaboration – Effectiveness



Large-scale application

- **<u>James Young and Todd Deutsch, NREL:</u>** PEC characterization and large-scale testing

- **<u>Tadashi Ogitsu, LLNL:</u>** Ab initio modeling of electrochemical interfaces

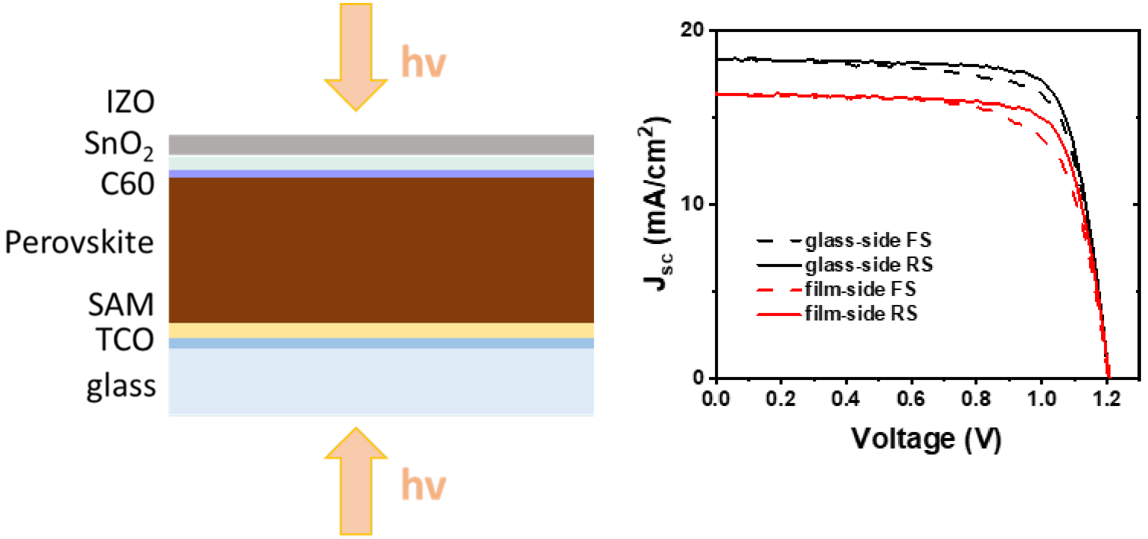
- **<u>Kai Zhu, NREL:</u>** Perovskite materials

- **<u>Myles Steiner, NREL:</u>** III-V tandem cells

- **<u>Joel Ager, LBNL:</u>** Photoelectrochemical AFM and STM

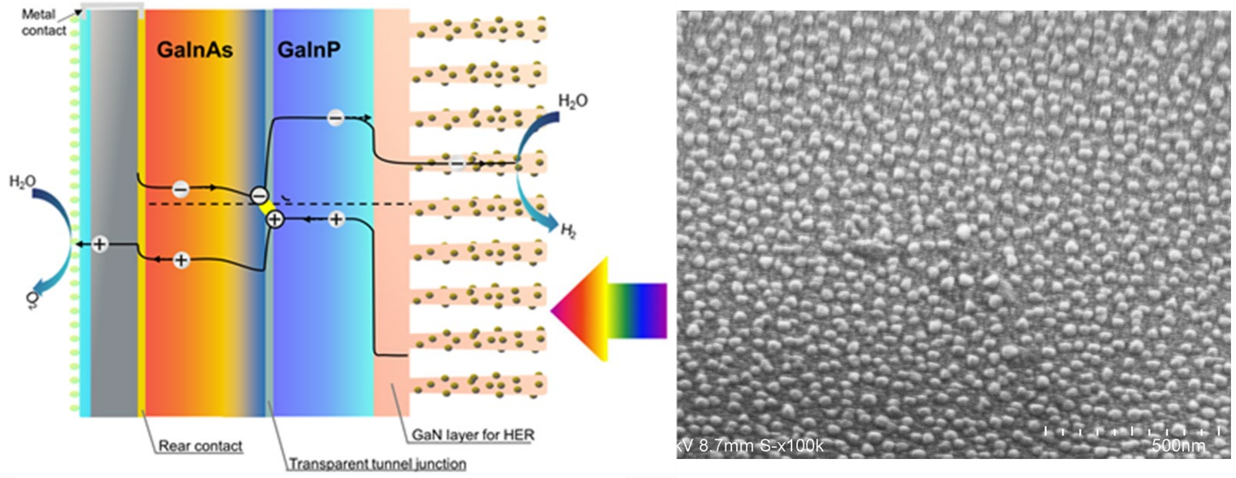
Collaboration – Effectiveness

Zhu (NREL), Yan (Toledo), Bhattacharya and Mi (UMich) co-designed and fabricated GaN-protected perovskite devices. We have established effective collaboration and achieved very promising results.



Schematic structure and performance of Perovskite solar cell

Steiner, James and Deutsch (NREL), Bhattacharya and Mi (UMich) have worked together to integrate GaN surface protection layer on III-V tandem devices and achieved very



Schematic of the device design (left) and SEM image of GaN nanostructures grown on III-V tandem device (right)

PI and their students attended PEC benchmarking/protocol workshop held in ASU in Sept. 2023, and plans to attend the workshop to be held in June 2024.

DEIA/Community Benefits Plans and Activities

- **This project has trained two female students, including Ms. Front Wongnonthawitthaya (undergraduate) and Ms. Huiyun Yang (master student).**

- **We also established connection with Prof. Jasmina Burek at UMass Lowell, a minority serving institution, for the life cycle assessment of solar water splitting devices and systems.**

Front Wongnonthawitthaya

Huiyun Yang

Proposed Future Work

Budget Period 2 (M13-24)

- Further improve the design, synthesis, integration, and performance of GaN-protected tandem photoelectrodes.
- Achieve GaN-protected perovskite/Si tandem photoelectrodes with STH of > 15% and maintaining 80% of the initial STH for >200 h.
- Achieve GaN-protected GaInP/GaInAs photoelectrode with STH >15% and maintaining 80% of the initial STH >250 h.

Budget Period 3 (M25-36)

- Demonstrate GaN-protected tandem device with a STH > 20% and stability of maintaining 80% of the initial STH for >500 h.
- Establish a GaN-protected PEC system to achieve hydrogen production at a rate of 0.1 g/h for diurnal operation for two weeks.

Technology Transfer Activities

- **No new IP has been generated since the start of this project.**

- **Previous IP on GaN-based solar water splitting in Mi's lab at the University of Michigan has been licensed to NX Fuels, Inc., which is a spin off company from the University of Michigan co-founded by Mi. NX Fuels is in the process of raising Series A funding for pilot scale demonstration and commercialization.**

Conflict of Interest: The University of Michigan and Mi have a financial interest in NX Fuels, Inc.

Project Summary

- **This project is focused on the development of GaN-protected high efficiency PEC tandem water splitting devices, with major innovations including:**
  - **The use of GaN nanostructures to protect against photocorrosion and oxidation and achieve long-term stable operation without compromising the device performance, thereby fundamentally addressing the outstanding stability issues in PEC water splitting.**
  - **The use of concentrated sunlight to significantly reduce the amount of materials used and therefore the overall system cost for solar hydrogen production.**
- **Major achievements to date:**
  - **Demonstrated the feasibility of low-temperature deposition of GaN on perovskite and III-V materials.**
  - **Demonstrated the effectiveness of using GaN nanostructures to protect the surface of III-V tandem devices, leading to relatively stable operation without significant performance degradation in 100 h.**
- **Success of this project will be instrumental to establish a low cost and scalable platform for high efficiency and highly stable PEC water splitting devices and systems by using industry ready materials, *e.g.,* perovskite, III-V, Si and GaN.**