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NATIONAL RENEWABLE ENERGY LABORATORY

Summary of NREL's FY13–FY15 Photovoltaic Subprogram

NREL Principal Investigators
National Renewable Energy Laboratory

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Technical Report
NREL/TP-5J00-68238
March 2017

Contract No. DE-AC36-08GO28308

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Introduction to NREL's FY13–FY15 Photovoltaic Subprogram

The U.S. Department of Energy (DOE) Solar Energy Technologies Office works to accelerate the market competitiveness of solar energy. The driving force behind this goal is the SunShot Initiative, a collaborative national effort that aggressively drives innovation to make solar energy fully cost competitive with conventional energy sources before 2020. The SunShot Initiative is designed to establish American technological and market leadership in solar energy, diversify the nation's electricity supply, reduce the environmental impacts of electricity generation, train the next generation of the solar workforce, and support broader national priorities, including national security, economic growth, and job creation.

Through SunShot, DOE supports efforts by businesses, non-profits, state and local governments, universities, and national laboratories to drive down the cost of utility-scale solar electricity to about 6 cents per kilowatt-hour and the cost of distributed solar electricity to at or below retail electricity rates. The national laboratories, including the National Renewable Energy Laboratory (NREL), are key players in the SunShot Initiative. Beginning in FY 2013, the national laboratories' SunShot activities were evaluated and funded by DOE on a three-year cycle and through a peer merit review process.

Through the SunShot collaboration with national laboratories in FY13–FY15, the Lab Proposal Development Process (LPDP), principal investigators at the national laboratories were invited to propose transformative ideas that address SunShot's goals and metrics. The ideas were submitted to DOE in the form of full proposals without the concept paper stage. Each full proposal was subjected to a rigorous review process, including evaluation by an external peer review panel consisting of distinguished scientists, engineers, and practitioners from academia, industry, and government laboratories across the nation. Applications that reviewed poorly were revised and then re-reviewed externally. After the review, the project milestones were negotiated, but a statement of project objective (SOPO) was not revised. Once funding awards were made, SunShot team members actively managed projects on a quarterly basis through the length of the award agreement to ensure that awardees met agreed-upon project objectives, delivered on milestones with valuable results, and remained relevant to the current research and development needs of the technology and the marketplace.

Proposals were solicited in spring of 2012 for the funding cycle lasting from FY13–FY15. The solicitation was open to multiple national laboratories, with submissions solicited in the following topical areas:

- Concentrating Solar Power
- Photovoltaics
- Soft Costs
- Systems Integration.

In this report, you will find summaries of the completed FY13–FY15 Photovoltaic projects that were funded within NREL. The summaries describe the initial motivation for the project; significant achievements, including publications, intellectual property, and collaborations; and remaining challenges. Among the NREL projects, you will find research of almost every major PV technology—from the next generation of silicon PV to relatively new organic PVs—as well as projects advancing PV module durability and characterization. Each of these projects was designed to support SunShot's goals, putting the United States one step closer to widespread use of low-cost, clean electricity. The last five projects were originally funded from the Systems Integration subprogram. But because of their direct relevance to PV, management of these was migrated into the Photovoltaic Subprogram during the latter part of this performance period. For completeness, therefore, these are included in this summary.

Sincerely,

Sarah Kurtz, Photovoltaic Subprogram Manager
Gregory Wilson, Photovoltaic Subprogram Manager, FY13–FY15
National Center for Photovoltaics, National Renewable Energy Laboratory

An Integrated Approach to Organic Photovoltaics

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Nikos Kopidakis (Primary Investigator), 303-384-6222, nikos.kopidakis@nrel.gov

Budget (FY13-FY15): $4.4M Agreement #: 25780

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1. Agreement Description and Motivation

The goal of this agreement was to advance Organic Photovoltaics (OPV) as a viable commercial PV technology. The scope of the program addressed all aspects of OPV: active materials development, contacts and device architectures and identification and mitigation of degradation mechanisms.

Over the past three years, we have fine-tuned an approach for developing new OPV polymer donor materials that combines expertise in electronic structure calculations, synthesis, and the characterization of photoinduced charge carrier dynamics. Our impact in the field using this approach can be summarized with several sets of systematic design rules developed for tuning material band gaps, energy levels, photoconductance, morphology, stability, etc. with different "R" groups, side chains, and backbones. Our publication record reflects and defines those systematic efforts.

Over the course of the program, we also developed several theoretical and experimental capabilities described in section 2.

During our program, we have partnered with industry from start-ups to international chemical and petroleum companies to further develop OPV materials and devices through funds-in Collaborative Research and Development Agreements (CRADAs). As such, we are able to move tools and technological advancements quickly to industrial partners, thus actively working to enable OPV as a commercial technology.

2. Significant Achievements

In this program we have created ground rules for designing, synthesizing and testing new OPV materials. Our main contribution is twofold: first, design rules for control and improvement of material properties and second practical structures for improved intrinsic photostability. As the search for higher efficiency and improved stability continues, we expect that our contributions will play a significant role in the development of a practical OPV technology. Our goal is for our unique materials database to serve as the starting point for materials discovery, our spectroscopic techniques to assist in material screening and optimization and our demonstrated stable materials to relax encapsulation requirements in device applications.

In the following we highlight specific achievements in our program.

2.1. Active-layer materials development.

- We developed a unique high-throughput combinatorial structure generation scheme. We combined building blocks (53 donors, 85 acceptors, 12 spacers, 20 functional groups) and computed using density functional theory (DFT) and time-dependent DFT (TDDFT) the properties of more than 26,000 molecules, extrapolating the results to predict electronic structure for more than 11,000 polymer structures (the manuscript describing the extrapolation method has been published in *J. Phys. Chem. C* **120**, 9650-9660 (2016)). .In total, the database has: 116,497 oligomer calculations and 42,487 extrapolated polymer structures.
- We developed an approach to calculating torsional potentials for practical conjugated polymer backbones used in push-pull copolymers that bypasses the requirement for expensive DFT calculations. Our method can compute the potential at a high level for both oligomers and as one approaches the polymer limit. The manuscript detailing our approach is currently in preparation.
- We used molecular dynamics simulations to predict the morphology of ~100,000 atom films for five different polymer systems (containing ~100 oligomers). These calculations allow for unprecedented visualization of the structure of polymer films.
- For snapshots taken from the MD simulation trajectories (previous bullet), we have extracted individual oligomers from the

simulations and computed their electronic structure. We find, for example, that the HOMO orbitals (which are what approximately the states that holes move through in these films) are highly mobile, migrating back and forth along the chain in response to slight shifts in inter-ring angles along the backbone. These motions have implications for how one should compute inter-chain hopping rates, with inter-chain couplings not well represented by any single snapshot, so they must be replaced by their time averaged values.

- Simulation Toolkit for Renewable Energy Advanced Molecular Modeling (STREAMM): The software we have used to do the above calculations has been packaged into a set of tools and scripts: STREAMM. The software has been approved for release and is available at: (github.com/NREL/streamm-tools.git) with documentation available at www.streamm.nrel.gov.
- With input from the database and from our chemical synthesis expertise, we designed and synthesized 12 new conjugated donor−acceptor (D−A) copolymers with a wide range of tunable band gaps employing ethynylene linkages to control steric interactions between neighboring repeat units in the backbone (*Macromolecules* **2013**, *46*, 3367-3375). This linking system also provided a synthetic route to copolymerize two electron-withdrawing monomers in alternating fashion, which allowed us to tune polymer donor energy levels much deeper than otherwise possible with those core molecules. We demonstrated that the photoconductance of such a polymer could be improved when matched and blended with a fullerene acceptor having appropriately deep energy levels, *i.e.,* $C60(CF_3)_2$.
- We reported a systematic study of well-defined alternating polymer structures with repeating (D−A), (D−D−A), and (D−D−D−A) units, and we compared the properties of those materials with those of five semi-random D−A copolymers with tunable D:A ratios (*ACS Macro Lett.* **2014**, *3*, 622−627). We demonstrated that the polymer donor LUMOs could be fine-tuned over a range of 0.4 eV, and we could use this set of materials to probe optimal energy level offsets with fullerene acceptors. We further demonstrated that the lifetimes of the photoconductance transients of copolymer:fullerene films were dramatically influenced by the D:A ratio in the copolymer. These results highlighted the power of this synthetic strategy for fine-tuning both the optoelectronic and photophysical properties of polymer donor materials.
- In 2014, we introduced a versatile, readily functionalized building block to the OPV community (cyclopenta[c]thiophene-4,6-dione, or CTD). We demonstrated that the choice of side-groups attached to this core had a very strong effect on morphology and photocarrier generation and recombination dynamics (*Adv. Energy Mater*. **2014**, 1301821). We were further able to correlate these effects on device performance. As also discussed below, the CTD unit is proving to be the most stable A-unit against photobleaching in D-A copolymers to date.
- In 2015, we better detailed the versatility of the CTD building block by comparing the properties of copolymers comprised of unsubstituted CTD with those of methyl, fluoro, and methyl cyano functionalized CTD, using those groups to dramatically tune optoelectronic energy levels (*J. Mater. Chem. A*, **2015**, *3*, 9777-9788).
- In the same paper, we also detailed our integrated approach toward the development of new narrow bandgap copolymers, which employs electronic structure calculations to guide the selection of target materials and uses time-resolved microwave conductivity (TRMC) to screen those materials for their photoconductance properties prior to their incorporation into complete OPV devices. This process can greatly accelerate materials development as it guides the time-consuming tasks of synthesis and device optimization to only focus on the most promising structures. Our methodology has attracted industry partnerships (most recently, a TSA with Phillips 66), interested in guiding and accelerating their materials development.

2.2. Contacts and device architectures

- We developed a Scanning Kelvin Probe microscopy methodology for the direct measurement of the electric field profile across the thickness s of an OPV device.
- We have shown the influence of contact materials on the electric field distribution in the active layer, which provides insight into the contact requirements for OPV. For example, we have shown that a molecular dipole monolayer affects the electric field distribution and magnitude in the active layer. This observation correlates to device measurements, where the optimized dipole

improves both FF and Voc. A manuscript detailing our findings has been published (*J. Phys. Chem. Lett.* **6**, 2269-2276 (2015)).

2.3. *Identification and mitigation of degradation mechanisms*

- We have undertaken a systematic study of the photobleaching of OPV active layers as unencapsulated films on quartz. We have identified unstable moieties in the backbone and side chains and we have synthesized variants of basic backbone structures to verify our hypothesis.
- We showed that the BDT-CTD copolymer, designed in this program, is the most stable OPV donor against photobleaching. A manuscript describing the findings of our combinatorial study of D-A copolymer stability as a function of the chemical structure of A is in preparation.
- Many state-of-the-art OPV active layers employ additives to improve performance. A typical such additive is diiodooctane (DIO). We carried out a study of the effect of DIO on the photostability of OPV polymers and showed that residual DIO in the active layer has an adverse effect on photostability, manifested as an increased rate of photobleaching. The manuscript describing these findings has been published (*Chem Mater.* ***28*** *876-884 (2016))*.
- For the above study, we developed, for the first time, a simple method to detect the presence of DIO in an organic film using X-ray fluorescence (XRF). We showed that when DIO is added to the solution, it is present in the final film. We also showed that DIO is removed from the film if the sample is placed in high vacuum (simulating the conditions for top contact evaporation). Our findings suggest that DIO must be avoided in roll-to-roll fabrication schemes since its presence in the film will increase degradation.

3. Modifications and Remaining Challenges

- One of the goals of our program going forward is to fully utilize the information in the materials database. We intend to make part of the existing database freely available to the OPV community as a resource for materials discovery. We expect that further concept structures will be designed by the community and included in future expansions of the database.
- We will explore (and add as needed) theoretical materials with non-optimum bandgap to enable transparent OPV for window applications. This is needed for BIPV, widely recognized to be the first entry of OPV into the energy generation market.
- Our wide array of characterization tools will greatly assist OPV development by providing high throughput screening of materials as well as information on device operation. We will use this toolset to evaluate high performance materials from literature and provide this information to the OPV community. Initial efforts on utilizing these tools by industry are ongoing (currently with Phillips 66) and our goal is to expand this to other companies as well.
- There is a strong need for the demonstration of improved OPV stability. In this program we developed one of the most stable OPV donors reported and work is ongoing to understand the molecular origin of this enhanced stability.
- The milestones that were not achieved are due to redirection of efforts within the subtasks. For example, efficiency prediction from theory (M8) was not pursued as the MD on the optimized films was only finished in FY15Q4. The latter is a major success of the effort and is one of the starting points for our FY16-17 work. Another milestone missed was the evaluation of T80 under normal and accelerated conditions (M25, 29 and 30). The majority of the effort on degradation was directed toward the intrinsic photostability of the active materials with remarkable progress (see above). Device degradation is currently being carried out in FY16-17 with the goal of identifying and understanding the relationship between molecular structure and device degradation.

4. FY13-FY15 Publications

- Z.R. Owczarczyk, W.A. Braunecker, A. Garcia, R.E. Larsen, N. Kopidakis, D.S. Ginley, and D.C. Olson, 5,10-Dihydroindolo[3,2- b]indole-Based Copolymers with Alternating Donor and Acceptor Moieties for Organic Photovoltaics. *Macromolecules* **46**, 1350–1360 (2013).
- W.A. Braunecker, S.D. Oosterhout, Z.R. Owczarczyk, R.E. Larsen, N. Kopidakis, D.S. Ginley, B.W. Larson, O.V. Boltalina, S.H. Strauss, and D.C. Olson, Ethynylene-Linked Donor–Acceptor Alternating Copolymers. *Macromolecules* 130429104833008 (2013). doi:10.1021/ma400238t

- Zbyslaw R. Owczarczyk, Wade A. Braunecker, Stefan D. Oosterhout, Nikos Kopidakis, Ross E. Larsen, David S. Ginley, and Dana C. Olson, Cyclopenta [c] thiophene-4, 6-dione-Based Copolymers as Organic Photovoltaic Donor Materials. *Adv. Energy Mater.* **4**, 1301821 (2014).
- Wade A. Braunecker, Stefan D. Oosterhout, Zbyslaw R. Owczarczyk, Nikos Kopidakis, Erin L. Ratcliff, David S. Ginley, and Dana C. Olson, Semi-random vs. Well-Defined Alternating Donor–Acceptor Copolymers. *ACS Macro Lett.* **3**, 622–627 (2014).
- Bradley A. MacLeod, Bertrand J. Tremolet de Villers, Philip Schulz, Paul F. Ndione, Hyungchul Kim, Anthony J. Giordano, Kai Zhu, Seth R. Marder, Samuel Graham, Joseph J. Berry, Antoine Kahn, and Dana C. Olson, Stability of inverted organic solar cells with ZnO contact layers deposited from precursor solutions. *Energy Environ. Sci.* **8**, 592–601 (2015).
- Jennifer L. Braid, Unsal Koldemir, Alan Sellinger, Reuben T. Collins, Thomas E. Furtak, and Dana C. Olson, Conjugated Phosphonic Acid Modified Zinc Oxide Electron Transport Layers for Improved Performance in Organic Solar Cells. *Acs Appl Mater. Interf.* **6**, 19229–19234 (2014).
- Stefan D. Oosterhout, Nikos Kopidakis, Zbyslaw R. Owczarczyk, Wade A. Braunecker, Ross E. Larsen, Erin L. Ratcliff, and Dana C. Olson, Integrating theory, synthesis, spectroscopy and device efficiency to design and characterize donor materials for organic photovoltaics: a case study including 12 donors. *Journal of Materials Chemistry A* **3**, 9777–9788 (2015).
- Unsal Koldemir, Jennifer L. Braid, Amanda Morgenstern, Mark Eberhart, Reuben T. Collins, Dana C. Olson, and Alan Sellinger, Molecular Design for Tuning Work Functions of Transparent Conducting Electrodes*, J. Phys. Chem. Lett.* **6**, 2269-2276 (2015)
- Ross E. Larsen, Simple extrapolation method to predict the electronic structure of conjugated polymers from calculations on oligomers, *J. Phys. Chem. C* **120**, 9650-9660 (2016).

- Removal of Residual Diiodooctane Improves the Photostability of a High-performance Organic Solar Cell Polymer, Bertrand J. Tremolet de Villers, Kathryn A. O'Hara, Dave P. Ostrowski, Perry H. Biddle, Michael L. Chabinyc, Sean E. Shaheen, Dana C. Olson, and Nikos Kopidakis, *Chem. Mater.* ***28*** *876-884 (2016)*
- *3 manuscripts in preparation*.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- 21 presentations at international conferences.
- ROI: p-type Doped Interlayer Using Polymeric Acid.
- ROI: Tunable 'Green Chemistry' Low Band Gap Conjugated Polymers Based Upon Functionalized Azine Linkages.
- Methods for producing thin film charge selective transport layers.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Solvay Fluorides	Hanover, Germany	Development of fluorinated donors for stable OPV	$1200
Solar Window Technologies (formerly New Energy Technologies)	John Conklin john@solarwindow.com	Development of mini-modules for BIPV	913
US Army NSRDEC		OPV textiles	800
Phillips 66	Woody, Kathy B Kathy.B.Woody@p66.com	Screening of OPV materials with TRMC	25

Rapid Development of Earth-abundant Thin Film Solar Cells Using Inverse Design

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Andriy Zakutayev (Primary Investigator), 303-384-6467, andriy.zakutayev@nrel.gov

Budget (FY13-FY15): $1.5M   Agreement #: 25785

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1. Agreement Description and Motivation

Diversification of future PV technology options requires a sustained effort in accelerated development of inorganic thin film solar cells. The goal of this project is to develop and demonstrate Rapid Development approach to new thin film solar cell prototypes made out of previously unknown materials. The first two tasks of the project aim at developing PV technologies based on relatively unknown ternary copper sulfides from Cu-Sn-S and Cu-Sb-S systems. The third task explores materials properties of Sn^{2+} oxides, which are completely unknown for PV absorber application. The final project milestone is to document the aim, methods, and examples of the Rapid Development approach in a peer-reviewed article, and demonstrate a 10% efficient device using a Cu-M-S absorber.

2. Significant Achievements

- Tested a novel approach to screening of materials for potential PV absorbers based on composition/elements, rather than traditional structure/stoichiometry. This led to down-selecting from ~10 Cu-Sn-S candidate absorbers to one promising Cu_2SnS_3 absorber through a combination of theory and experiment. [Appl. Phys. Lett., 103, 253902 (2013)].
- Experimentally demonstrated control of doping in Cu_2SnS_3 through both point defects and alloying with isostructural metallic Cu_3SnS_4 phase. These studies also have shown that Cu_2SnS_3 is not a line compound and tends to exist in cation-disordered structure. [Chem. Mater., 26, 4951 (2014)].
- Developed a theoretical approach to describe cation disorder in tetrahedrally-bonded solids and applied it to Cu_2SnS_3 and related materials. The calculations indicate that the cation disorder effects transport and modifies doping in Cu_2SnS_3, and allows predicting materials less prone to disorder. [Phys. Rev. Appl. 3, 034007 (2015)].
- Experimentally elucidates effects of cation disorder on carrier transport in Cu_2SnS_3 by controlling its cation ordering through annealing. Whereas the cation ordering significantly changes doping and majority charge carrier transport properties, it has minimal effect of the minority carrier transport. [Phys. Rev. Appl. 4, 044017 (2015)].
- Predicted the existence of extended anti-site defects in tetrahedrally bonded materials, which cannot be captured within a simple point defect model. Such extended defects in Cu_2SnS_3 have significantly lower formation energy and their formation may strongly affect materials optoelectronic properties. [Phys. Rev. B 92, 201204(R) (2015)].
- Established a self-regulated growth approach, by which the stoichiometric single-phase $CuSbS_2$ or $CuSbSe_2$ absorber is formed, whereas the excess Sb_2S_3 remains in the vapor phase. Within the self-regulated growth regime, $CuSbS_2$ doping can be controlled in 10^{16} -10^{18} cm^{-3} range by changing the substrate temperature and Sb_2S_3 flux. [Sol. En. Mat. Sol. Cells 132, 499 (2015)].
- Demonstrated a combinatorial approach to accelerated development of thin film photovoltaic device prototypes on the example of $CuSbS_2$ and $CuSbSe_2$ absorbers. This enables facile optimization of the performance as a function of the phase purity, crystallographic orientation, layer thickness and other absorber parameters. [Progress in Photovoltaics 24, 929, (2016)].
- Performed screening of the back contacts for the $CuSbS_2$ absorber using the combinatorial device development approach. Among many candidates, Mo/MoO_x back contact was found optimal for increasing device short circuit

current. [IEEE 42nd Photovoltaic Specialist Conference (PVSC), 2436 (2014)].

- Established and optimized the post-deposition thermal treatment (annealing) conditions in Sb_2S_3 vapor, which improves the $CuSbS_2$ absorber morphology, stress, photoconductivity, and photoluminescence characteristics. The absorber material improvements translate into increase of the fill factor and ideality factor of the PV devices. [J. Phys. Chem. C, 120, 18377 (2016)].
- Developed an alternative $CuSbQ_2$ (Q=S,Se) heterojunction partner with higher conduction band position, specifically (Cd,Zn)S:Ga deposited by ALD. These experimental results are based on the theoretical predications of the electron affinity as a function of Cd/Zn ratio. [Mater. Chem. Front. (2017), DOI: 10.1039/C6QM00291A].
- Translated the knowledge developed on the example of $CuSbS_2$ into $CuSbSe_2$ photovoltaic devices. The self-regulated growth and the CdS/Mo contacts on $CuSbSe_2$ absorbers led to solar cells with 3% efficiency. [Appl. Phys. Exp. 8, 082301 (2015)].
- Subsequent optimization of the $CuSbSe_2$ deposition process lead to nearly 5% efficiencies and relative trade offs in the solar cell performance. The photoexcited charge carrier collection in $CuSbSe_2$ solar cells is enhanced by drift in an electric field increasing J_{sc}, which also limits the V_{oc} of the resulting photovoltaic devices. [Advanced Energy Materials DOI: 10.1002/aenm.201601935, (2017)].
- Documented the motivation, goals, methods of the "Rapid Development" approach in an invited review-type article on advanced materials for solar energy conversion. The paper also includes the discussion of the advances needed in science and technology to meet the outstanding challenges in this area. [Journal of Optics, 18, 073004 (2016)].
- Published a review paper on defects and disorder in multinary tetrahedrally bonded semiconductors. These general phenomena are exemplified by the aforementioned work on the Cu-Sn-S absorbers. [Semicond. Sci. Technol. 31 123004 (2016)].
- Theoretically predicted the possibility of tuning optical absorption of SnO by isovalent alloying with Mg, Ca, Sr and Zn. Suitable band gaps and optical properties close to that of direct semiconductors are achievable, while the comparatively small effective masses are preserved in the SnO alloys. [APL Mater. 4, 106103 (2016)].
- Experimental synthesis and characterization results support the feasibility of the (Sn,Zn)O alloy growth. The $Sn_{1-x}Zn_xO$ thin films showed an increase in the absorption coefficient in the range from 1 eV to 2 eV, which is consistent with the theoretical predictions for the isovalent alloying. [Chemistry of Materials 28, 7765, (2016)].

3. Modifications and Remaining Challenges

- To address the unexpected challenges related to cation disorder that came up in the Cu_2SnS_3 part of the project, a theoretical model for disorder in tetrahedrally bonded materials was created. As a result of the modeling, a decision was made to redirect in year-3 the resources from Cu_2SnS_3 to $CuSbQ_2$ absorber materials. Beyond the Cu_2SnS_3 work, this model has much broader applications to predicting effects of disorder in other tetrahedral materials.
- It is likely that 1 of the 22 milestones will not be met by the end of the project. Specifically, the 10% PV device efficiency using a novel Cu-M-S absorber will probably not be achieved – mostly due to a somewhat unexpected need for a complete device/contacts re-design due to different α/m^* balance and different CB position of $CuSbS_2$ compared to CIGS or CZTS. However, note that the 10% was set intentionally as a stretch goal at the beginning of this project, since no other inorganic thin film PV technology showed such rapid progress from 0% to 10% in the past. Also, note that a significant progress towards addressing these challenges was achieved, such as Sb_2S_3 vapor annealing, (Cd,Zn)S:Ga heterojunction partners, and MoO_x back contacts - this led to nearly 5% efficient $CuSbSe_2$ PV devices which should still attract a significant interest in the thin film PV community. So overall, despite the one missed milestone, this work allowed us to establish the unique "Rapid Development" approach, meeting the goal of the project (see above). In the future, this "Rapid Development" approach can now be applied to other more promising novel PV absorber technologies.

4. FY13-FY15 Publications

- Evaluation of photovoltaic materials within the Cu-Sn-S family, P. Zawadzki, L. L. Baranowski, H. Peng, E. S. Toberer, D. S. Ginley, W. Tumas, A. Zakutayev, and S. Lany, Appl. Phys. Lett., 103, 253902 (2013).
- Control of doping in Cu2SnS3 through defects and alloying L. L. Baranowski , P.P Zawadzki , S. T. Christensen , D. Nordlund , S. Lany , A. C. Tamboli , L. Gedvilas , D.S. Ginley , W. Tumas , E. S. Toberer , A. Zakutayev, Chem. Mater., 26, 4951 (2014).
- Entropy-driven clustering in tetrahedrally bonded multinary materials, P.P. Zawadzki, A. Zakutayev, S. Lany, Phys. Rev. Appl. 3, 034007 (2015).
- Effects of disorder on carrier transport in Cu_2SnS_3, L. L. Baranowski, K. McLaughlin, P. Zawadzki, S. Lany, A. Norman, H. Hempel, T. Unold, E. S. Toberer, and A. Zakutayev, Phys. Rev. Appl. 4, 044017 (2015).
- Extended anti-site defects in tetrahedrally bonded semiconductors, P. Zawadzki, A. Zakutayev, and S. Lany, Phys. Rev. B 92, 201204(R) (2015)
- Comparison of Cu_2SnS_3 and $CuSbS_2$ as Potential Solar Cell Absorbers, A. Zakutayev, L. L. Baranowski, A. W. Welch, C. A. Wolden, and E.S. Toberer, Proceedings of the IEEE 40th Photovoltaic Specialist Conference (PVSC), 2436 (2014).
- Self-regulated growth and tunable properties of $CuSbS_2$ solar absorbers, A. W. Welch, P. P. Zawadzki, S. Lany, C. A. Wolden, A. Zakutayev, Solar Energy Materials and Solar Cells 132, 499 (2015).
- Copper antimony chalcogenide thin film PV device development, A. W. Welch, Lauryn L. Baranowski, F. W. de Souza Lucasa, E. S. Toberer, C.A. Wolden, A. Zakutayev, Proceedings of the IEEE 42nd Photovoltaic Specialist Conference (PVSC), 2436 (2014).
- Thermal treatment improvement of $CuSbS_2$ absorbers, F. W. de Souza Lucas, A. W. Welch, L. L. Baranowski, P. C. Dippo, L.H. Mascaro, A. Zakutayev, Proceedings of the IEEE 42nd Photovoltaic Specialist Conference (PVSC), 2436 (2014).
- Effects of Thermochemical Treatment on $CuSbS_2$ Photovoltaic Absorber Quality and Solar Cell Reproducibility, F. W. de Souza Lucas, A. W. Welch, L. L. Baranowski, P. C. Dippo, P.C., H. Hempel, T. Unold, R. Eichberger, B. Blank, U. Rau, L. H. Mascaro, and A. Zakutayev, J. Phys. Chem. C, 120, 18377 (2016).
- Accelerated development of $CuSbS_2$ thin film photovoltaic device prototypes, A. W. Welch, L. L. Baranowski, P. Zawadzki, C.DeHart, S. Johnston, S. Lany, C. A. Wolden, and A. Zakutayev, Progress in Photovoltaics 24, 929, (2016).
- Conduction band position tuning and Ga-doping in (Cd,Zn)S alloy thin films, L. L. Baranowski, S. Christensen, A. W. Welch, S. Lany, M. Young, E. S. Toberer, A. Zakutayev, Mater. Chem. Front. (2017), DOI: 10.1039/C6QM00291A.
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- Pathway to oxide photovoltaics via band-structure engineering of SnO, H.Peng, A. Bikowski, A. Zakutayev, and S. Lany, APL Mater. 4 , 106103 (2016).
- Advanced materials for solar energy conversion, in “Roadmap on optical energy conversion”, S. Lany, T. Gershon and A. Zakutayev et al, Journal of Optics, 18, 073004 (2016).
- A review of defects and disorder in multinary tetrahedrally bonded semiconductors, L. L Baranowski, P. Zawadzki, S. Lany, E.S. Toberer and A.Zakutayev, Semicond. Sci. Technol. 31 123004 (2016).
- Synthesis and characterization of (Sn,Zn)O alloys, A. Bikowski, A. Holder, H.Peng, S. Siol, A. Norman, S. Lany, and A. Zakutayev, Chemistry of Materials 28, 7765, (2016)
- Trade-Offs in Thin Film Solar Cells with Layered Chalcostibite Photovoltaic Absorbers, A. W. Welch, L. L. Baranowski, H. Peng, H. Hempel, R. Eichberger, T. Unold, Stephan Lany, C. Wolden, A. Zakutayev (2017), Advanced Energy Materials, DOI: 10.1002/aenm.201601935.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- Adam Welch, Presentation highlight, MRS fall meeting, 2013.
- Lauryn Baranowski, Best Presentation Award, Conference on Earth and Energy Research.
- Lauryn Baranowski, Student Presentation Award, Symposium E, MRS spring meeting, 2014.

- Pawel Zawadzki, Postdoc Presentation Award, Symposium E, MRS spring meeting, 2014.
- Lauryn Baranowski, Best Poster Nomination, Symposium V, MRS fall meeting, 2014.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Colorado School of Mines	manit@asu.edu	Assistance in synthesis, characterization and analysis of Cu-M-S materials and devices through graduate student subcontracts	200

Multifunctional Transparent Conducting Oxides

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: David Ginley (Primary Investigator), 303-384-6573
david.ginley@nrel.gov

John Perkins (Co-Primary Investigator), 303-384-6606
john.perkins@nrel.gov

Budget (FY13-FY15): $1.5M Agreement #: 25776

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1. Agreement Description and Motivation

Transparent conductors (TCs), particularly transparent conducting oxides (TCOs), are enabling contacts for current and emerging photovoltaics. Historically, TC materials have been selected from a very small basis set: ZnO-, SnO_2-, and In_2O_3-based materials. However, as TC applications and thereby their requirements diversify, so do needs for improved materials, processes and opto-electronic characteristics. These may include electron affinity, process temperature, environmental stability, surface chemistry, and processability. Overlaid on all of these issues is the drive to use low-cost, earth-abundant materials. Clearly one solution cannot fit all (i.e., CZTS, Si, and OPV devices have different requirements). The purpose of this activity is to build a set of TCO solutions that can be employed in PV technologies to improve the efficiency, manufacturability, cost and reliability of photovoltaic devices.

2. Significant Achievements

- Identified Zn vacancies as the dominant compensating defect in Ga-doped Zn(Mg)O and approach to mitigate them.
- Demonstrated conductivity σ > 1000 S/cm for sputtered TiO_2 based TCOs on glass.
- Demonstrated conductivity σ > 1000 S/cm for sputtered SnO_2 based TCOs on glass.
- Transferred Ga-doped ZnMgO deposition process from PLD on single crystal substrates to large area sputtering on glass.
- Demonstrated atmospheric pressure processed silver-nanowire / zinc-oxide (AgNW/ZnO) composite TC with $R_s \approx 10$ ohms/sq. and T ≈ 85 %.
- Demonstrated low-stress, high-conductivity a-InZnO TCO thin films by sputtering.
- Developed new Kinetically Limited Mobility (KLM) theoretical model for amorphous mixed metal oxide TCOs.
- Held industry/academia/laboratory workshop with industry, academia and labs, which confirmed an ongoing need for improved TC materials., a report was published.
- The program graduated 3 PhD candidates over the three years.

3. Modifications and Remaining Challenges

- At the end of Year 2, the focus of this Agreement was changed at DOE's request from a materials research focus on more device relevancy. This resulted in every original Year 3 milestone being dropped and 5 completely new Year 3 milestones were developed. Some of the Year 3 Milestones were not met because it was realized that one component, the device testing of ZnOS buffer layers on CIGS devices was not realizable within the Year 3 resources of this project.

4. FY13-FY15 Publications

Journal Publications

- "Confirmation of the dominant defect mechanism in amorphous In-Zn-O through the application of in situ Brouwer analysis", S.L. Moffitt, A.U. Adler, T.O. Mason, T. Gennett, D.S. Ginley, and J.D. Perkins, J. American Ceramic Society, **98**, 2099, doi:10.1111/jace.13518. (2015).
- "Multivalency of Group 15 Dopants in SnO2", H. Peng, J.D. Perkins, and S. Lany, Chemistry of Materials, **26**, 4876, doi:10.1021/cm502411g. (2014).
- "Modeling amorphous thin films: Kinetically limited minimization", P. Zawadzki, J.D. Perkins, and S. Lany, Physical Review B, **90**,

094203, doi:10.1103/PhysRevB.90.094203. (2014).

- "Processing-phase diagrams: a new tool for solution-deposited thin-film development applied to the In5O(OPri)13–In2O3 system", R.M. Pasquarelli, M.F.A.M. van Hest, P.A. Parilla, J.D. Perkins, R. O'Hayre, and D.S. Ginley, Journal of Materials Chemistry C, **2**, 2360, doi:10.1039/c3tc31930j. (2014).
- "Enhanced Electron Mobility Due to Dopant-Defect Pairing in Conductive ZnMgO", Y. Ke, S. Lany, J.J. Berry, J.D. Perkins, P.A. Parilla, A. Zakutayev, T. Ohno, R. O'Hayre, and D.S. Ginley, Advanced Functional Materials, **24**, 2875, doi:10.1002/adfm.201303204. (2014).
- "Reactive sputtering of amorphous In-Zn-O TCO from metallic targets", J.D. Perkins, M. Nix, A.A. Dameron, A. Zakutayev, T. Gennett, and D.S. Ginley, (2013). doi:10.1109/pvsc.2013.6744347.
- "Optimizing amorphous indium zinc oxide film growth for low residual stress and high electrical conductivity", M. Kumar, A.K. Sigdel, T. Gennett, J.J. Berry, J.D. Perkins, D.S. Ginley, and C.E. Packard, Applied Surface Science, **283**, 65, doi:10.1016/j.apsusc.2013.06.019. (2013).
- "Non-equilibrium origin of high electrical conductivity in gallium zinc oxide thin films", A. Zakutayev, N.H. Perry, T.O. Mason, D.S. Ginley, and S. Lany, Applied Physics Letters, **103**, 232106, doi:10.1063/1.4841355. (2013).

Conference Proceedings

"Opportunities for Improving Photovoltaic Performance with Better Transparent Contacts", D.S. Ginley, and J.D. Perkins, Proceedings of the 42nd IEEE Photovoltaic Specialists Conference (PVSC), New Orleans, LA (2015).

- "Atmospheric Pressure Processed Silver-Nanowire (Ag-NW) / ZnO Composite Transparent Conducting Contacts", J.D. Perkins, S. Aggarwal, M.F.A.M. van Hest, T. Gennett, and D.S. Ginley, Proceedings of the 42nd IEEE Photovoltaic Specialists Conference (PVSC), New Orleans, LA (2015).
- "Improving mechanical stability and electrical properties of silver nanowire films with a zinc tin oxide overcoat", S. Aggarwal, M.F.A.M. van Hest, J.D. Perkins, and D.S. Ginley, Proceedings of the 40th IEEE Photovoltaic Specialist Conference (PVSC), Denver, CO (2014).
- "Improving Electron Transport in Ga-doped Zn0.7Mg0.3O, a Wide-Gap Band-Edge-Energy-Tunable Transparent Conducting Oxide", J.D. Perkins, Y. Ke, S. Lany, J.J. Berry, A. Zakutayev, B. Gorman, T. Ohno, P.A. Parilla, R. O'Hayre, and D.S. Ginley, Proceedings of the 40th IEEE Photovoltaic Specialist Conference (PVSC), Denver, CO (2014).
- "Reactive sputtering of amorphous In-Zn-O TCO from metallic targets", T. Gennett, J.D. Perkins, M. Nix, A.A. Dameron, and D.S. Ginley, Proceedings of the SPIE Optics and Photonics Conference, San Diego, CA (2013).
- "Reactive sputtering of amorphous In-Zn-O TCO from metallic targets", J.D. Perkins, M. Nix, A.A. Dameron, A. Zakutayev, T. Gennett, and D.S. Ginley, Proceedings of the 2013 IEEE 39th Photovoltaic Specialists Conference (PVSC), (2013), doi:10.1109/pvsc.2013.6744347.
- "Reactive sputtering of amorphous In-Zn-O TCO from metallic targets", J.D. Perkins, M. Nix, A.A. Dameron, A. Zakutayev, T. Gennett, and D.S. Ginley, Proceedings of the 2013 IEEE 39th Photovoltaic Specialists Conference (PVSC), (2013), doi:10.1109/pvsc.2013.6744347.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- Yi Ke, MRS Graduate Student Award - Silver Medal, Materials Research Society Dec. 2013.
- “Conformal Coating of Highly Structured Surfaces”, D. Ginley, J. Perkins, J. Berry and T. Gennett, U.S. Patent # 8329502, Awarded 12/11/2012.
- "Using Amorphous Zinc-Tin Oxide Alloys in the Emitter Structure of CISG PV Devices", P. Hersh, M. van Hest, D. Ginley, J. Perkins, V. Bollinger, Application # 20140020744, Submitted 1/3/2013.
- “Methodology for Improved Adhesion for Deposited Fluorinated Transparent Conducting Oxide Films on a Substrate”, T. Gennett, J. Perkins, A. Dameron, Application 14/542,448. Submitted 11/14/2014.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Prof. Ryan O'Hayre Colorado School of Mines	rohayre@mines.edu	Electronic Doping of Tunable Band Gap Transparent Conductors	**180**

Perovskites Effort within the PV Director's Initiative

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Joseph J. Berry (Primary Investigator), 303-384-7611, joe.berry@nrel.gov

Budget (FY13-FY15): $850K Agreement #: 25786

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1. Agreement Description and Motivation

The purpose of this agreement was to perform an examination of the technological hurdles to the development of hybrid perovskite solar cell technologies. This single year agreement (FY15) was centered on the three key elements of any deployable photovoltaic (PV) technology, namely efficiency, stability and scalability. These three elements are required to calculate the key metric of the SunShot program for PV, namely the levelized cost of electricity (LCOE). The efforts were designed to cast a relatively broad assessment across these three elements of the most conventional hybrid-perovskite systems

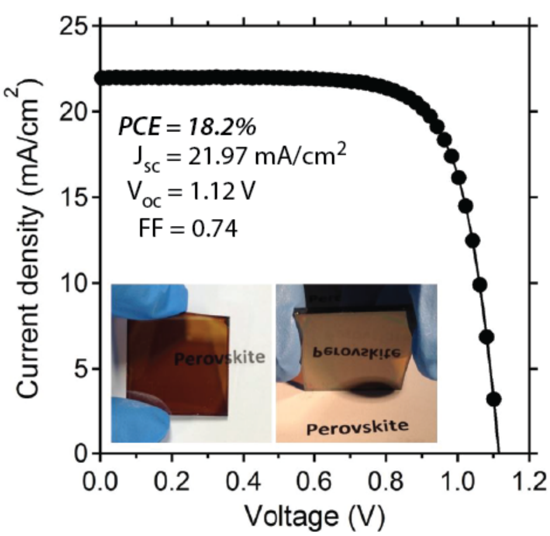

Fig. 1. Data for NREL champion device (as of 2/20/2015). Characterization was performed as per Snaith et. al.'s approach and reported value is a stabilized power conversion efficiency (PCE). Device uses a F:SnO_2/TiO_2/PAL/spiro-MeOTAD/Ag planar architecture. Lower panels show typical perovskite active layer films.

based on methylammonium lead iodide ($MAPbI_3$). This program was able to identify several critical aspects related to the stability of these devices, with detailed studies of both the active layer and contact interfaces. With respect to the active layer, addressing stability appears to be a

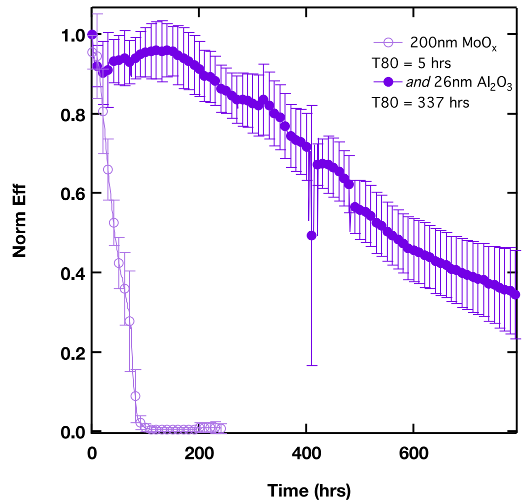

Fig. 2. Data displaying normalized power conversion efficiency vs. time. Each point is the PCE as determined by J-V analysis for a: F:SnO_2/TiO_2/PAL/spiro-MeOTAD/MoO_x/Al device stack and the same stack with an additional 2nm Al_2O_3 overlayer.

combination of equal parts kinetics and thermodynamics. The interfacial aspects have been elucidated with unique NREL capabilities to provide insight into mechanisms at play in stable device configurations reported in the literature. Devices based on input from these interfacial studies will be undertaken within this program moving forward. In addition, thermodynamic stability will be addressed with several approaches to alter the active layer stability via substitution of different organic components for MA, specifically formamidinium and cesium. The kinetic aspect is more complex but is being addressed via improved synthetic approaches undertaken by the larger community (including other work supported by SunShot), which the program has leveraged. The final milestone to demonstrate a device with PCE>15% and an improvement of greater than 2x in stability from a standard control devices was achieved as shown in Fig 1 and Fig 2. These were done for the conventional $MAPbI_3$ material with the alternative alloys to be investigated in FY16-18.

2. Significant Achievements (FY15)

There were myriad technical achievements for this program in its brief period of performance (FY15 only). In addition to establishing internal baselines for work on perovskite photovoltaics (i.e. performance levels and processing conditions), the team has been able to begin to differentiate the work supported by EERE in this program from other work being done within the community. Specifically, efforts in this year provided insight into important aspect of device level performance via the use of cross-sectional scanning Kelvin probe techniques applied to perovskite photovoltaics devices coupled to use of surface and interface science studies. These experiments provide insight into the operational principles of cells based on these materials. In particular results indicate that the details of the device architecture appear to vary depending on the fabrication details. NREL work demonstrated that efficient devices can be produced which exhibit p-n heterojunctions (in contrast to previously reported p-i-n heterojunctions). There has also been considerable work to examine the role of interfaces in both device performance and stability. This project was able to confirm that devices based on perovskite active layers at lab scale compare favorably to other thin film technologies with internal champion devices exceeding the 15% PCE targeted as the year end milestone. Device studies demonstrated that the current collection and by extension, material homogeneity and/or design of contact for these devices is clearly the primary limiter of performance with respect to PCE. This is further confirmed by the interfacial studies conducted by NREL. The role these considerations play in the stability of these device systems is the current focus of work moving forward. The ability to reproducibly create high performance PALs and devices is critical to these assessments, which are required to ultimately enable efficient modules. Work was also performed to examine roadblocks for this technology as it pertains to scaling of these devices.

- Contributed to international efforts for defining challenges in hybrid-perovskite materials research with 5 publications submitted (3 accepted and 2 still in the review process) as detailed in the publications section. In addition there are a number of draft publications circulating within the team, which are anticipated to be submitted within the first half of FY16.
- Invited presentation at the NanoGe:SPINS15 conference in Spain reporting on several aspects of stability in perovskite materials and devices.
- Invited talk to SPIE-OPV15 on improved processing for PSCs.

3. Modifications and Remaining Challenges

- Based on the results of the initial work in these materials, stability remains a challenge. Although a greater than 2x improvement over baseline devices was realized this degradation of first generation $MAPbI_3$ device is very rapid in ambient conditions under operation. While there are demonstrations in the literature of more robust shelf life of these systems it is not obvious these are technologically relevant. The more limited device operation studies have shown promise that technologically relevant operational lifetime targets can be achieved. There remains a clear need to understand the mechanism by which this is achieved so that these results can be applied to higher performance and more easily scalable systems.
- Device and materials reproducibility also appears to be a challenge for the community and even across laboratory spaces within the NREL program. While our primary lab space in the FTLB routinely produces cells with PCE>15%, out labs in the SERF result in devices more in the 12% range. Despite some initial internal round-robins this has continued to be a problem with some closing of the gap with the addition of new postdoctoral researchers.
- The programmatic prioritization on understanding the mechanisms of stability and the associated analytic measurements rather than exclusively device measurements stability assessments reduced output of some of the device level stability assessments. Although other programs (organic photovoltaics) were generous with resources for these tests, bandwidth for these studies was somewhat limited. This should be largely addressed with the new solar parameter analysis (SPA) SPA system moving into FY16-18.
- The rate of advance in this field is such that details of approach (e.g. examination of specific device architectures) became largely irrelevant in comparison to more foundational

analytic efforts. As a result there were a number of progress indicators that were at less than 100% completion, largely as a result of loss in technical merit relative to the Year 1 milestone and progress in the field. Opportunities for other avenues to advance perovskite PV and the need to shift direction also played a role. Given the program milestone to demonstrate devices with PCE of over 15% and to demonstrate an improvement in the time to 80% of initial PCE of 2x under constant load, there were a number of progress indicators while relevant initially not required to meet these targets. More importantly these indicators as initially conceived no longer represented novel approaches to provide a foundation for enabling the more aggressive targets required for this technology to succeed in the future. Specifically the development of inverted device architectures was not completed along with the associated progress indicators (1.2 tasks 1.2.1 and 1.2.2). This was also done with consideration for the existing literature. Instead work to gain insight into the more fundamental aspect of alternative HTM configurations was undertaken (e.g. NiOx and other oxide on top of PAL, and carbon-nanotube on top of PAL interface studies) with device studies not yet completed (i.e. reported in peer-reviewed journals) due to the more novel nature and expected impact of these experiments and devices. These studies were complemented by studies of Spiro-OMeTAD the canonical HTM to understand its deficiencies more quantitatively, and there by circumvent them. This resulted in knock-on effects to task 3.1 and progress indicator 3.1.3 specifically, with completion of this anticipated early in FY16. This along with existing obligations for device degradation tools also pushed completion of associated tasks (2.2 tasks, progress indicator 2.2.4) further out. Given the expansion of device stability testing tools (i.e. SPA) and the presence of numerous groups working on these inverted geometries, we anticipate that there will be opportunities to evaluate these device architectures via partnerships with other research groups and potentially industry. The final component task that remained incomplete (4.1 task 4.1.1 and 4.1.2) related to scaling of PSC devices. Although the team has demonstrated larger area (1 cm^2 devices) above 12% these have been produced via spin coating. In addition, devices of greater than 15 cm^2 were also demonstrated by the team, again using spin coating techniques. Initial devices using scalable production were not, however, completed. The team was able to produce ETLs on TCO via a scalable processing approach and anticipate completion of initial PAL via spray processing shortly after the end of this reporting period resulting in minimal impact on work moving into the next funding cycle (FY16-18).

- Based on these issues and to move successfully into the next program cycle, the SETO and NREL staff worked closely in the construction of this new program's SOPO. This collaboration has produced a resulting SOPO with progress indicators and milestones that are constructed in a more robust, realistic and technically oriented fashion.

4. FY15 Publications

- J. Berry, T. Buonassisi, D. A. Egger, G. Hodes, L. Kronik, Y.-L. Loo, I. Lubomirsky, S. R. Marder, Y. Mastai, J. S. Miller, D. B. Mitzi, Y. Paz, A. M. Rappe, I. Riess, B. Rybtchinski, O. Stafsudd, V. Stevanovic, M. F. Toney, D. Zitoun, A. Kahn, D. Ginley, and D. Cahen, "Hybrid Organic-Inorganic Perovskites (HOIPs): Opportunities and Challenges," *Adv. Mater.*, Jul. 2015.
- C. Jiang, M. Yang, Y. Zhou, B. To, S. Nanayakkara, J. Luther, W. Zhou, J. Berry, J. van de Lagemaat, N. Padture, K. Zhu, M. Al-Jassim "Nanometer-Scale Profiling of Electrical Potential in Perovskite Solar Cells: Carrier Separation and Transport." *Nature Communications* (Accepted 8/18/2015)
- Philip Schulz, Anne-Marie Dowgiallo, Mengjin Yang, Kai Zhu, Jeffrey Blackburn and Joseph J. Berry "Charge Transfer Dynamics between Carbon Nanotubes and Hybrid Organic Metal Halide Perovskite Films" *Advanced Materials* (under review)
- Chuanxiao Xiao, Zhen Li, Harvey Guthrey, John Moseley, Ye Yang, Sarah Wozny, Helio Moutinho, Bobby To, Joseph J. Berry, Brian Gorman, Yanfa Yan, Kai Zhu, and Mowafak Al-Jassim. "Mechanisms of electron-beam-induced damage in perovskite thin films revealed by cathodoluminescence spectroscopy." *J. Phys. Chem. Lett.* (under review)
- P. F. Ndione, W.-J. Yin, K. Zhu, S.-H. Wei, and J. J. Berry. "Avenue to Controlling the Stability of Organometallic Perovskite Thin

Films" (Accepted, 9/16/2015) *J. Mater. Chem. A*

5. FY15 Special Recognitions, Awards, and Patents:

Although there has been work on development of IP, at this point none of the developed ROIs have moved to the patent process.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Sean Saheen University of Colorado at Boulder	Sean.Shaheen@Colorado.edu	Device modeling, characterization and student support	70
Alan Sellinger Colorado School of Mines	aselli@mines.edu	Synthesis of surface/interface modifiers	45

Low-Cost, High-Efficiency, One-Sun HVPE III-V Solar Cells

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Aaron Ptak and David Young (Primary Investigators), 303-384-6660, aaron.ptak@nrel.gov

Budget (FY13-FY15): $1.5M Agreement #: 25781

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1. Agreement Description and Motivation

During this program, NREL established a research program aimed at drastically reducing the costs of III-V solar cells, with an overall goal of viable III-V devices in one-sun and low-concentration applications. To be successful, the costs associated with both high-quality epitaxy and the single-crystal substrates must be addressed. Toward that end, we realized the design, construction, and operation of a custom dual-chamber HVPE growth system that has demonstrated much lower material costs and much higher throughput than current-generation III-V growth technologies.

In parallel, we initiated research into controlled spalling for the removal of devices from a parent wafer by mechanical fracture with the goal of allowing the wafer to be reused multiple times for substantial cost reduction. Controlled spalling is an innovative substrate removal process that cleaves a wafer parallel to the substrate surface at a precise depth. This process is extremely fast and inherently low cost, requiring only a cheap, electroplated strained metal layer deposited on the film.

The final milestone for this agreement is:
Grow, process and measure a GaAs solar cell grown at >1 μm/min with conversion efficiency >23%; Determine minimum and maximum spalling facet excursions vs. Ni stressor thickness and electroplating current density. Demonstrate maximum peak-to-valley facet heights <10 μm over an area >1 cm^2 at an average spall depth of 8.5±1.0 μm.

2. Significant Achievements

- Designed and constructed a custom, automated, dual chamber HVPE reactor, with significant aid from the NREL Engineering Team, and put in place all safety systems and procedures.
- Produced GaAs with high optoelectronic quality at growth rates exceeding 1.8 μm/min.
- Demonstrated that our dual chamber design produces flat, parallel, low-defect homo- and hetero-interfaces.
- Demonstrated GaAs homojunction devices, without passivating layers, with conversion efficiency >18%; demonstrated >30% increase in J_{sc} with the addition of an InGaP window layer.
- Developed predictive computational fluid dynamics model of the NREL HVPE reactor.
- Established relationships between spall depth and Ni stressor thickness for (100) GaAs, enabling tunable average spall depth.
- Identified multiple engineering factors that control spalling facet height, allowing {110} facet height to be reduced to <10 μm.
- Spalled active devices and measured their performance, with cells showing no appreciable degradation.

3. Modifications and Remaining Challenges

The original vision for this new project was the development of high quality III-V layers in an HVPE system that would be built as part of the project, plus demonstration of a large number of substrate reuses via controlled spalling. This project was funded at the "seed" level as part of the Type II Core section of the original call for proposals.

Early on, it was realized that construction of the HVPE system at NREL would take much longer than the plan that was based on previous design/build experience with a university subcontractor. The system was highly engineered for performance and safety, but did not come online until ~17 months into the project. This meant scaling back the timeline to reach the desired efficiencies of >25%. The eventual final project milestone included a 23% efficiency metric, which was missed.

The HVPE reactor produced excellent material quality very soon after start-up and looked to be capable of reaching the desired efficiencies by the end of the project. However, a leak in the gas line plumbing caused significant contamination of the reactor, and the system was inoperative for ~10 months during maintenance and cleaning, leading to a high level of schedule slippage. Much of the reactor has been repaired, and contamination levels are orders of magnitude lower than previously observed, allowing the system to again produce high-quality material. Currently, efficiencies of ~18% have been achieved for growth rates >1 μm/min. We expect that continued contamination mitigation and further development of HVPE growth will enable conversion efficiencies >20% in the next year.

The new effort in controlled spalling was expected to lead to 100's of substrate reuses during this project. It was determined that repeating the same procedure multiple times was an inefficient use of project resources, so the decision between NREL and DOE was made to focus on understanding the spalling process and understanding the limits of the technique on (100) GaAs.

The final outcome of the controlled spalling aspect of this project is that spall depth in (100)-oriented materials can be accurately controlled, and that {110} facets can be confined to a region <10 μm. We feel that further refinement of this technique may decrease the extent of the facets by up to a factor of two. Given current cost/μm estimates for HVPE growth, an order of magnitude reduction in the necessary spall layer thickness may be required and is unlikely to be achievable in (100) GaAs due to its unavoidable crystallographic faceting. Previously unknown sensitivities of controlled spalling to velocity, method of force application, and edge effects discovered in this project are informing work on (100) Ge, which does not exhibit faceting. Therefore, we have decided to cease work in this area in favor of other promising liftoff technologies. The controlled spalling part of the final project milestone was achieved.

4. FY13-FY15 Publications

- K.L. Schulte, A.W. Wood, R.C. Reedy, S.E. Babcock, A.J. Ptak, and T.F. Kuech, Heteroepitaxy of GaAs on (001) ⇒6° Ge substrates at high growth rates by HVPE, J. Appl. Phys. 113, 174903 (2013). http://dx.doi.org/10.1063/1.4803037
- J. Simon, D. Young, and A. Ptak, Low Cost III-V Solar Cells Grown by Hydride Vapor Phase Epitaxy, Proceedings of the 40th IEEE Photovoltaic Specialist Conference (PVSC) 538 (2014). http://dx.doi.org/10.1109/PVSC.2014.6925127
- C.A. Sweet, J.D. Simon, D.L. Young, A.J. Ptak, and C.E. Packard, Understanding the factors that control spalling fracture in single crystal semiconductors: Progress towards wafer reuse, submitted to J. Appl. Phys. (2014).
- C.A. Sweet, J.D. Simon, D.L. Young, A.J. Ptak, and C.E. Packard, Effect of Material Choice on Spalling Fracture Parameters to Exfoliate Thin PV Devices, Proceedings of the 40th IEEE Photovoltaic Specialist Conference (PVSC) 1189 (2014). http://dx.doi.org/10.1109/PVSC.2014.6925127
- K.L. Schulte, J. Simon, D.L. Young, and A.J. Ptak, Modeling of Gas Curtains in a Dual Chamber Hydride Vapor Phase Epitaxial Photovoltaic Growth Reactor, 42nd IEEE Photovoltaic Specialist Conference (PVSC) (2015).
- J. Simon, K.L. Schulte, D.L. Young, and A.J. Ptak, GaAs Solar Cells Grown by Hydride Vapor Phase Epitaxy and the Development of GaInP Cladding Layers, submitted to J. Photovoltaics (2015).
- C.A. Sweet, J.E. McNeely, B. Gorman, D.L. Young, Engineering controlled spalling in (100)-oriented GaAs for wafer reuse, A.J. Ptak, and C.E. Packard, 42nd IEEE Photovoltaic Specialist Conference (PVSC) (2015).
- K.L. Schulte, J. Simon, A.Roy, D.L. Young, T.F. Kuech, and A. J. Ptak, Computational fluid dynamics-aided design of a hydride vapor phase epitaxy reactor optimized for photovoltaic devices, submitted to J. Cryst. Growth (2015).

5. FY13-FY15 Special Recognitions, Awards, and Patents

- "High Throughput Semiconductor Deposition System", D.L. Young, A.J. Ptak, T.F. Kuech, K. Schulte, and J. Simon, U.S. Patent Application SN 14/801,551, NREL ROI 12-47, filed May 2013.

- C. Sweet, 1st and 2nd place awards for Best Scientific Art, Colorado Center for Advanced Ceramics Conference, August 2013.
- A. Ptak, Research Participant Program Outstanding Mentor Award, NREL, August 2013.
- J. Simon, *et al.*, Best Poster Award, Photovoltaics Specialists Conference (PVSC), June 2014.
- C. Sweet, *et al.*, Best Poster Award runner-up, Photovoltaics Specialists Conference (PVSC), June 2014.
- HVPE Team, NREL President's Award, for the design, construction, and implementation of a Hydride Vapor Phase Epitaxy reactor capable of producing low-cost, high-quality photovoltaic devices at very high growth rates, September 2014.
- J. Simon, NREL Technology Transfer Office Rising Star Award, February 2015.
- K. Schulte, *et al.*, Best Poster Award, Photovoltaics Specialists Conference (PVSC), June 2015.
- C. Sweet, *et al.*, Best Poster Award runner-up, Photovoltaics Specialists Conference (PVSC), June 2015.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Colorado School of Mines Corinne Packard	cpackard@mines.edu	Controlled film delamination and wafer reuse for thin film GaAs photovoltaics	164.956

III-V Multijunction Concentrator Solar Cells

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: John Geisz (Primary Investigator), 303-384-6474, john.geisz@nrel.gov

Budget (FY13-FY15): $6.8M Agreement #: 25775

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1. Agreement Description and Motivation

The focus of this agreement was the *development of a 48%-efficient four-junction inverted metamorphic solar cell (4J IMM)* designed for real-world concentrating photovoltaic (CPV) operating conditions. This advance over pre-existing ~43%-efficient three-junction solar cells has the potential to provide close to a 10% decrease in the resulting levelized cost of electricity (LCOE), significantly accelerating the achievement of the SunShot goal. Perhaps even more important is *enabling science-based advances in understanding of the metamorphic (i.e. plastically relaxed) materials and multijunction device physics* that this work will produce. Specifically, we attempted to understand and control 1) dislocation formation and glide in materials for III-V multijunction cells; 2) cell design, material choices, and growth techniques for device elements such as tunnel junctions in the presence of dislocations; and 3) electrical and optical coupling between individual junctions within complex multijunction solar cells. These challenging scientific advances were *required* to achieve these high efficiency device goals, and produced a published body of foundational knowledge that serves as a platform for the multijunction field to build upon for years to come. The final milestone of the project is a 48%-efficient 4J IMM solar cell.

2. Significant Achievements

- Developed metamorphic grade to InP lattice-constant with threading dislocation densities < 3×10^6 cm^{-2}.
- Identified and described mechanism for enhanced dislocation glide in CuPt-ordered grades, as well as a new mechanism for dislocation formation.
- Demonstrated metamorphic solar cell junctions spanning band-gaps of 1.4 eV to 0.70 eV with W_{oc} < 0.4 V
- Developed novel metamorphic $Ga_{0.76}In_{0.24}As$ / $GaAs_{0.75}Sb_{0.25}$ tunnel junction for use between third and fourth junctions.
- Demonstrated 4J IMM concentrator solar cell with confirmed efficiency of 45.7% at 234 suns concentration, 45.6% at 690 suns, and 45.2% at 1000 suns.
- Demonstrated 4J IMM one-sun efficiency of 39.8%.
- Advanced the understanding of multijunction solar cell physics and experimental techniques to quantify the implications of light emission on device performance. (Luminescent coupling , radiative efficiency, voltage-independent photoluminescence, etc.)
- Advanced the accuracy and understanding of multijunction solar cell measurements. (Luminescent coupling effects on EQE, subcell characterization through electroluminescence and selectively varied illumination IV measurements, importance of spectral adjustment, etc.)
- Developed methods for estimating energy yield of complex multijunction solar cells including effects of luminescent coupling. We have also shown how the cells can be redesigned to optimize performance accounting for luminescent coupling.

3. Modifications and Remaining Challenges

- Improvements in the accuracy of multijunction concentrator solar cells throughout and as a result of this project has reduced the systematic overestimation of efficiencies. Specifically, spectral control of concentrator flash measurements for more than two junctions (using T-HIPSS) and recognition of area definition uncertainties has made achieving the 48% milestone more challenging. If our correctly-measured 46% efficiency solar cells were measured using previous techniques, the resulting incorrect efficiency would have been overestimated to be approximately 48%.

- As a result of selective wet chemical etching of the mesa area, the areas of different junctions may be considerably different resulting in uncertainties in subcell current densities. This makes controllable current matching adjustments very challenging. We have begun to define the area using a processed aperture with initial success, but find it to be technically challenging for the thicker 4J IMM. Dry etching capabilities that could improve mesa definition are also planned for future years.
- Tech transfer of the technology developed here to commercial production is underway with an established partner through funds-in CRADA.

4. FY13-FY15 Publications

Peer-Reviewed Journals

- W. E. McMahon, J. Kang, R. M. France, A. G. Norman, D. J. Friedman and S. H. Wei, "Ordering-enhanced dislocation glide in III-V alloys." J Appl Phys 114, p. 203506 (2013). http://dx.doi.org/10.1063/1.4833244
- D. J. Friedman, J. F. Geisz and M. A. Steiner, "Analysis of Multijunction Solar Cell Current–Voltage Characteristics in the Presence of Luminescent Coupling." IEEE Journal of Photovoltaics 3, p. 1429 (2013). http://dx.doi.org/10.1109/JPHOTOV.2013.2275189
- M. A. Steiner, J. F. Geisz, I. Garcia, D. J. Friedman, A. Duda, W. J. Olavarria, M. Young, D. Kuciauskas and S. R. Kurtz, "Effects of Internal Luminescence and Internal Optics on Voc and Jsc of III-V Solar Cells." IEEE Journal of Photovoltaics 3, p. 1437 (2013). http://dx.doi.org/10.1109/JPHOTOV.2013.2278666
- D. J. Friedman, J. F. Geisz and M. A. Steiner, "Effect of Luminescent Coupling on the Optimal Design of Multijunction Solar Cells." IEEE Journal of Photovoltaics 4, p. 986 (2014). http://dx.doi.org/10.1109/JPHOTOV.2014.2308722
- R. M. France, I. Garcia, W. E. McMahon, A. G. Norman, J. Simon, J. F. Geisz, D. J. Friedman and M. J. Romero, "Lattice-Mismatched 0.7-eV GaInAs Solar Cells Grown on GaAs Using GaInP Compositionally Graded Buffers." IEEE Journal of Photovoltaics 4, p. 190 (2014). http://dx.doi.org/10.1109/JPHOTOV.2013.2281724
- R. M. France, W. E. McMahon, J. Kang, M. A. Steiner and J. F. Geisz, "In situ measurement of CuPt alloy ordering using strain anisotropy." J Appl Phys 115, p. 053502 (2014). http://dx.doi.org/10.1063/1.4863821
- I. Garcia, R. M. France, J. F. Geisz and J. Simon, "Thin, high quality GaInP compositionally graded buffer layers grown at high growth rates for metamorphic III-V solar cell applications." J Cryst Growth 393, p. 64 (2014). http://dx.doi.org/10.1016/j.jcrysgro.2013.10.043
- I. Garcia, J. F. Geisz, R. M. France, J. Kang, S.-H. Wei, M. Ochoa and D. J. Friedman, "Metamorphic $Ga_{0.76}In_{0.24}As$ / $GaAs_{0.75}Sb_{0.25}$ tunnel junctions grown on GaAs substrates." Journal of Applied Physics 116, p. 074508 (2014). http://dx.doi.org/10.1063/1.4892773
- M. P. Lumb, M. A. Steiner, J. F. Geisz and R. J. Walters, "Incorporating photon recycling into the analytical drift-diffusion model of high efficiency solar cells." J Appl Phys 116, p. 194504 (2014). http://dx.doi.org/10.1063/1.4902320
- J. S. Ward, A. Duda, D. J. Friedman, J. F. Geisz, W. E. McMahon and M. Young, "High aspect ratio electrodeposited Ni/Au contacts for GaAs-based III-V concentrator solar cells." Prog. Photovolt: Res. Appl. 23, p. 646 (2015). http://dx.doi.org/10.1002/pip.2490
- I. García, W. E. McMahon, M. A. Steiner, J. F. Geisz, A. Habte and D. J. Friedman, "Optimization of multijunction solar cells through indoor energy yield measurements." IEEE Journal of Photovoltaics 5, p. 438 (2015). http://dx.doi.org/10.1109/JPHOTOV.2014.2364128
- J. F. Geisz, M. A. Steiner, I. García, R. M. France, D. J. Friedman and S. R. Kurtz, "Implications of Redesigned, High-Radiative-Efficiency GaInP Junctions on III-V Multijunction Concentrator Solar Cells." IEEE Journal of Photovoltaics 5, p. 418 (2015). http://dx.doi.org/10.1109/JPHOTOV.2014.2361014
- R. M. France, J. F. Geisz, I. García, M. A. Steiner, W. E. McMahon, D. J. Friedman, T. Moriarty, C. R. Osterwald, J. S. Ward, A. Duda, M. Young and W. Olavarria, "Quadruple junction inverted metamorphic concentrator devices." IEEE Journal of Photovoltaics 5, p. 432 (2015).

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- J. F. Geisz, M. A. Steiner, I. Garcia, R. M. France, W. E. McMahon and D. J. Friedman, "Generalized Optoelectronic model of series-connected multijunction solar cells." IEEE Journal of Photovoltaics, 5, p. 1827, (2015). http://dx.doi.org/10.1109/JPHOTOV.2015.2478072
- J.S. Park, J. Kang, B. Huang, S. H. Wei and W. E. McMahon, "Period-doubling reconstructions of semiconductor partial dislocations." NPG Asia Materials, 7, p. e216, (2015). http://dx.doi.org/10.1038/am.2015.102
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- I. Garcia, R. M. France, J. F. Geisz, W. E. McMahon, M. A. Steiner, S. Johnson and D. J. Friedman, "Metamorphic III-V solar cells: recent progress and potential." IEEE Journal of Photovoltaics, 6, p. 266, (2015). http://dx.doi.org/10.1109/JPHOTOV.2015.2501722
- M. Ochoa, M. A. Steiner, I. Garcia, J. F. Geisz, D. J. Friedman and C. Algora, "Influence of temperature on luminescent coupling and material quality evaluation in inverted lattice-mismatched and metamorphic multijunction solar cells." Prog. Photovolt: Res. Appl., 24, p. 357, (2015). http://dx.doi.org/10.1002/pip.2714
- R. M. France, W. E. McMahon and H. L. Guthrey, "Critical thickness of atomically ordered III-V alloys." Applied Physics Letters 107, p. 151903, (2015). http://dx.doi.org/10.1063/1.4933092
- J.S. Park, J. Kang, J.H. Yang, W.E. McMahon, S.H. Wei, "Polymerization of defect states at dislocation cores in InAs.", Journal of Applied Physics, 119, p. 045706, (2015). http://dx.doi.org/10.1063/1.4940743

Trade Journals

- Matthew Lumb, Robert Walters, Myles Steiner, and John Geisz, "Improving modeling of high-efficiency solar cells". Compound Semiconductor, 20, p. 62 (2014). http://www.compoundsemiconductor.net/article/95947-improving-modeling-of-high-efficiency-solar-cells.html

Conference Proceedings

- A. G. Norman, R. M. France, W. E. McMahon, J. F. Geisz and M. J. Romero, "The influence of atomic ordering on strain relaxation during the growth of metamorphic solar cells." J. Phys.: Conf. Ser. 471, p. 012006 (2013). http://dx.doi.org/10.1088/1742-6596/471/1/012006
- M. A. Steiner, J. F. Geisz, I. García, D. J. Friedman, S. R. Kurtz, "Experimental and modeling analysis of internal luminescence in III-V solar cells." AIP conference proceedings 1556, p. 57 (2014). http://dx.doi.org/10.1063/1.4822199
- I. Garcia, J. F. Geisz, R. M. France, M. A. Steiner and D. J. Friedman, "Component integration strategies in metamorphic 4-junction III-V concentrator solar cells." AIP conference proceedings 1616, p. 41 (2014). http://dx.doi.org/10.1063/1.4897024
- J. F. Geisz, R. M. France, I. Garcia, M. A. Steiner and D. J. Friedman, "Device characterization for design optimization of 4 junction inverted metamorphic concentrator solar cells." AIP conference proceedings 1616, p. 114 (2014). http://dx.doi.org/10.1063/1.4897041
- M. P. Lumb, M. A. Steiner, J. F. Geisz and R. J. Walters, "Analytical modeling of III-V solar cells close to the fundamental limit." Proceedings of SPIE 8981, p. 898114 (2014). http://dx.doi.org/10.1117/12.2041359
- J. F. Geisz, "Energy Yield Determination of Concentrator Solar Cells Using Laboratory Measurements." AIP conference proceedings 1679, Aix-les-Bains, (2015). http://dx.doi.org/10.1063/1.4931516
- I. Garcia, "Field Spectra Binning for Energy Yield Calculations and Solar Cell Design." Proceedings of the 42nd IEEE PVSC, New Orleans, (2015). https://doi.org/10.1109/PVSC.2015.7356207
- M. A. Steiner, "Radiation effects on luminescent coupling in III-V solar cells." Proceedings of the 42nd IEEE PVSC, New

Orleans, (2015). https://doi.org/10.1109/PVSC.2015.7355672

- T. Moriarty, R. France and M. Steiner, "Rapid, enhanced IV characterization of multi-junction PV devices under one sun at NREL." Proceedings of the 42nd IEEE PVSC, New Orleans, (2015). https://doi.org/10.1109/PVSC.2015.7355845

5. FY13-FY15 Special Recognitions, Awards, and Patents

- John Geisz, Alliance for Sustainable Energy, Chairman's Award for Exceptional Performance, September 2013.
- Ryan France, 42nd IEEE PVSC Outstanding Technical Contribution, June 2015.
- John Geisz, invited plenary speaker at 17th International Conference on Metalorganic Vapor Phase Epitaxy, July 2014.
- Andrew Norman, invited speaker at Microscopy of Semiconducting Materials Meeting, April 2013.
- Ryan France, invited speaker at the Lawrence Symposium on Epitaxy, February, 2014.
- Ryan France, invited speaker at Ohio State University, December 2014.
- Myles Steiner, invited speaker at SuperSolar, UK, Advances in Concentrator Photovoltaics, March 2015.

6. University and Industry Partners

The III-V Multijunction Concentrator Solar Cells project did not collaborate with any external industrial or university partners.

Next Generation Silicon PV

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Paul Stradins (Primary Investigator), 303-384-6774, pauls.stradins@nrel.gov

David Young (Co-Primary Investigator), 303-384-6621, david.young@nrel.gov

Budget (FY13-FY15): $8.686M Agreement #: 25783

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1. Agreement Description and Motivation

This Agreement addresses two ways to principally advance the dominant Silicon PV technology [1] to the next level. One, increase the module efficiency (and thus, the cell efficiency) above the current average 16-19% to 22-24% achieved in laboratories or few companies, while keeping the manufacturing cost close, or lower than, the current mainstream relatively low-efficiency Si PV. Two, in the longer ~15 years term, introduce cost-efficient Si tandems with efficiencies ~30% considerably above practical limits of the best Si cells. In both cases, the value of module efficiency (~6 ¢/W in module price per absolute efficiency point [2]) drives the total installation and levelized cost of electricity. These two critical issues for the Si PV are addressed by the two tasks (Task 1 and Task 2) of this project, emphasizing the cell efficiency and potentially industrial relevant ways to achieve it.

Task 1 focused on developing novel, high efficiency, industrially relevant, single junction n-Cz Si cells. Our principal goal is to develop up to 23% small area Si cell (4 cm^2) and adapt this knowledge to industrially relevant 156 mm, 22% baseline n-type Cz Si solar cell with passivated contacts. All the recent ultrahigh efficiency Si cells ~ 25% (Fraunhofer ISE [3], SunPower [4,37], Panasonic [5], Sharp [38]) are based on passivated contacts. FhISE cells use expensive high-end laboratory processes on n-FZ wafers not compatible with industry. While our cell process is not yet fully industry-compatible, it demonstrates innovative steps (e.g. passivated contacts and their metallization, bulk defect mitigation) directly on a n-Cz Si cell. In addition to project milestone goals, Task 1 established high efficiency, industrially relevant Si PV research program and technology base at NREL. This includes tools, processes, knowledge base, and collaborations with leading academia (FhISE, GIT) and industry (AMAT, Suniva, GTAT, SunEdison). The Task 1 cells also serve as bottom cells for Task 2.

Task 2 focused on high efficiency tandem development interfacing Si bottom cells with III-V top cells. The goal is to achieve > 25% efficient tandem cells using ~ 20% efficient Si bottom cells with high-quality III-V top cells of ~ 1.7 eV gap. Three parallel approaches were used: 1) new method to grow low-defect GaP buffer layer on Si (as interconnecting buffer to III-V cell); novel wafer bonding for Si/III-V tandem; 2) ultrahigh efficiency, mechanically stacked Si/III-V tandem. These were made possible by integration and joint effort of both Si and III-V teams. In addition to achieving milestones, this integration is a beneficial and unique outcome of this project, further enhancing the Si PV program at NREL.

The final milestone for this agreement is: "Demonstrate 22% on 156mmX156mm n-Cz solar and a GaInP/Si mechanically stacked 4-terminal cell with greater than 26% efficiency".

2. Significant Achievements

- Developed a new Si PV program at NREL, abandoning former low-efficiency thin film Si approach in favor of high efficiency, industrially relevant approach.
- Within 3 years, created the Si PV technical capability at NREL, estimated one of the 3 places in US that have it (NREL, GIT, ASU), especially clean Si processing capability that preserves ~ 5 ms bulk lifetime and >20% cells.
- Developed a tunneling passivated contacts approach based on poly-Si/SiO2 and TCO/SiO_2 first in the US, with GIT following and learning from us. Reached state of the art in this field, currently leading it together with FhISE, ISFH, and GIT. Best result: n-type passivated BSF contact to n-Cz wafer (Prov. patent application).

- Developed an original technique to further passivate the passivated poly-Si/SiO2 contact (Prov. patent application).
- Demonstrated state of the art, passivated contact to n-Cz Si wafer: (champion cell at iVoc= 735 mV, J_0 = 2.1 fA/cm^2 for n-type contact and iVoc= 690 mV, J_0 = 29 fA/cm^2 for p-type contact).
- Demonstrated high performance diffused, deep passivated B emitter to n-Cz Si wafer: iVoc= 717 mV, J_0 = 17 fA/cm^2, enabled by Tabula Rasa treatment and state of the art Al_2O_3/SiN_x passivation/ARC layer stack.
- Demonstrated 20% PERT cell on n-Cz wafer with deep passivated B emitter and patterned passivated P-diffused BSF.
- Demonstrated state of the art, high voltage, passivated contact n-Cz Si cells before metallization (champion cell at i-Voc= 724 mV with total J_0 = 12 fA/cm^2 and bulk lifetime 4.1 ms). This would enable 23% n-Cz cells with non-damaging Al metallization.
- Together with III-V team, demonstrated 27% III-V /Si tandem, over 26% expected by the Agreement final milestone.
- Introduced Tabula Rasa process to enable high efficiency n-type Si PV, originally not in the AOP plan. (Prov. patent application).
- Teamed with GIT and Fraunhofer ISE and won FPACE-II to complement the AOP.
- Demonstrated high quality ion-implanted passivated contacts in collaboration with Applied Materials, not in the original AOP.
- Developed deep DFT theory-based insights into electronic and structural properties of $Si_x(III\text{-}V)_y$ and $Si_x(II\text{-}VI)_y$ metastable alloys lattice-matched to Si.
- Developed a novel, TCO-mediated wafer bonding process for III-V/Si tandem. (ROI 15-17).
- Developed, together with III-V team, novel, Si-epitaxy free process to obtain single domain Si surfaces and practically defect-free MOCVD epitaxy of GaP on these surfaces.
- Established the following Si PV capabilities at NREL: Clean dopant diffusions and oxidations for 4” wafers; State of the art ALD Al_2O_3 and PECVD SiN_x passivation compatible with 23% cell efficiency; Clean Tabula Rasa process in diffusion and optical furnaces; High quality ~ 1.5 nm tunneling SiO_2 for passivated contact (both by thermal and wet chemistry); State of the art PECVD depositions of intrinsic, n-, and p-type a-Si:H on tunneling SiO_2; Ag and Cu grid lithography and electrodeposition; NiSi barriers for Cu diffusion; New Si cleanroom construction with 6” automated wet bench from Singulus and automated diffusion furnaces from Thermco.

3. Modifications and Remaining Challenges

- We could not reach the desired milestone Si cell efficiencies (22%) on large area, due to the lack of full size Si wafer tools at NREL, and significant delay in cleanroom construction into 2016. Our mitigation strategies were the following:
  a) subcontracted GIT for large area implanted B emitters/P-BSF to passivate them at NREL. This approach led to 20.1%, 156 mm cells, screen-printed at GIT.
  b) However, to reach milestone 22%, passivated back contact and plated contacts were needed. This includes 156 mm custom wet chemistries at NREL (Piranha, RCA, HNO_3).
  c) Collaborated with UNSW. They provided large area, diffused B emitters. However, BSF passivated contacts on 156 mm wafers could not be formed at NREL due to the lack of clean 156 mm annealing tools.
  d) Designed and ordered a single 156 mm annealing tube. Its construction was delayed by the Allen Scientific, to be delivered mid-September 2015.
  e) Designed and fabricated custom wet-processing baths for 5 wafers/bath.
  f) The above efforts will enable us to have limited 156 mm capability before the cleanroom is online, but are not sufficient to reach our 22% 156 mm milestone.
- Up to now (mid-September 2015) we experience significant delays in reaching target efficiencies (22% in FY14, and 23% in FY15) of small area Si cells. This is due to lack of clean, Si-dedicated metallization tools at NREL. They are too expensive to purchase with program funds, so we are currently refurbishing an old e-beam evaporator for Si project, and relying on NREL’s II-VI team and GIT for clean metallization. However, with our new Al-Si/Al/Ni/plated Ag grid approach, we are cautiously optimistic about reaching 22% in FY15.
- Since Q2 of FY14, several modifications were agreed and implemented to Task 2:
  1) exploration of novel wafer bonding for Si/III-V tandem;

2) mechanically stacked 26% Si/III-V tandem with III-V team's InGaP top cell;
3) phasing out of the Si-(III-V) alloy work because both experiment and theory strongly suggested difficulties in widening the optical gap above 1.3 eV.

- We did successfully develop Cu contact barrier layers based on NiSi, as well as Cu electrodeposition on those barriers. However, to achieve the milestone 22-23% Si cell efficiencies, we choose the Al-based contacts instead of Cu, because of the metal-induced degradation issues. Thus, the Cu barrier milestone is resource-limited (one student + part engineer project), leading to its delay beyond the end of FY15.
- The Si-(III-V) epitaxial alloy work was wrapped up in mid- FY14 in favor of wafer bonding and mechanical stacking approaches to tandem. The DFT theory was especially helpful to make this decision early, by demonstrating the principal difficulty to open the optical gap >1.3 eV due to Si-Si clustering.

Positive gains due to the above challenges:

- The bulk lifetime degradation, emphasized by lack of clean Si tools at NREL, helped us to develop the Tabula Rasa oxygen precipitate dissolution process (see Achievements).
- Because of the lack of Si-dedicated metallization tools, we gained insights of why the high efficiency passivated contact cells are so sensitive to metallization issues, and developed novel ways (capping interlayers) to mitigate it. Moreover, the lack of clean metallization tools forced us to concentrate on Al metal instead of Ag for cell contacts, an industrially relevant approach that enables impurity guttering.
- Initially we missed milestones for epitaxial GaP on Si, because we did not have proper Si MBE capabilities for Si pre-epitaxy. This forced us to develop a new, better GaP on Si epitaxy process with AsH_3 by MOCVD that does not involve Si pre-epitaxy.
- Because of the delay with GaP on Si epitaxy, we turned to a new approach to III-V /Si tandem (mechanic stack) and reached 27% tandem in half a year. This enhanced our visibility in the field and we have respected collaborations on this topic (ISFH and EPFL).

4. FY13-FY15 Publications

- Adele C. Tamboli, Maikel F. A. M. van Hest, Myles A. Steiner, Stephanie Essig, Emmett E. Perl, Andrew G. Norman, Nick Bosco, and Paul Stradins, "III-V/Si wafer bonding using transparent, conductive oxide interlayers", Appl. Phys. Lett. **106**, 263904 (2015); http://dx.doi.org/10.1063/1.4923444

- Emily L. Warren, Alan E. Kibbler, Ryan M. France, Andrew G. Norman, Paul Stradins, and William E. McMahon, "Growth of antiphase-domain-free GaP on Si substrates by metalorganic chemical vapor deposition using an in situ AsH3 surface preparation", Applied Physics Letters 107 , 082109 (2015) http://dx.doi.org/10.1063/1.4929714

- David L. Young, William Nemeth, Sachit Grover, Andrew Norman, Hao-Chih Yuan, Benjamin G. Lee, Vincenzo LaSalvia, Paul Stradins, "Carrier selective, passivated contacts for high efficiency silicon solar cells based on transparent conducting oxides", Energy Procedia 55 (2014) 733-740, http://dx.doi.org/10.1016/j.egypro.2014.08.053

- Sachit Grover, Jian V. Li, David L. Young, Paul Stradins and Howard M. Branz, "Reformulation of solar cell physics to facilitate experimental separation of recombination pathways", Appl. Phys. Lett. 103, 093502 (2013); http://dx.doi.org/10.1063/1.4819728

- Benjamin G. Lee, Shuo Lib, Guillaume von Gastrow, Marko Yli-Koski, Hele Savin, Ville Malinec, Jarmo Skarp, Sukgeun Choi, Howard M. Branz, "Excellent passivation and low reflectivity with atomic layer deposited bilayer coatings for n-type silicon solar cells", Thin Solid Films, v. 550, 1 January 2014, pp. 541–544 http://dx.doi.org/10.1016/j.tsf.2013.10.166

- William Nemeth, Matthew Page, Vincenzo LaSalvia, Steve Johnston, Robert Reedy, David L. Young, Pauls Stradins, "Materials Aspects of Polycrystalline Silicon Passivated Tunneling Contacts for High Efficiency Silicon Solar Cells", Journal of Materials Research, Manuscript JMR-2015-0637 currently under revision (2015).

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5. FY13-FY15 Special Recognitions, Awards, and Patents

- NREL Director’s award to three Si team members (Matt Page, Hao-Chih Yuan, William Nemeth) for developing Si PV capability at NREL (2013).
- Alliance President’s Award to Vincenzo LaSalvia for development of Tabula Rasa process for Si PV (2014).
- Two invited talks (2014) on our Si program achievement topics: WCPEC-6 (Japan) and AVS (Baltimore) – Stradins.
- Best Poster Award, IEEE PVSC 2015 – Tamboli.
- NCPV Fellowship for PhD student Rohan Chaukulkar (CSM) for a 9-month project to reveal physics of Si surface passivation by ALD Al_2O_3 (Aug. 2013 - April 2014).
- LDRD, FPACE-II, NextGen III awarded, synergistic to this project (Tamboli, Lee, Stradins, Warren).
- Three provisional patents (Passivated contacts, Tabula Rasa, Passivation of the Passivated Contact):
  1. “PECVD Polycrystalline Silicon Thin Film on Tunneling Silicon Oxide on Wafer Silicon”, ROI 14-59, Prov/app. under negotiation with NREL Tech Transfer.
  2. NREL PROV/14-79 “Optical Annealing Treatment For High Efficiency Solar Cell Wafers” (2014) No. 62/110,289;
  3. NREL PROV/15-51 “Hydrogenation of Passivated Contacts” (2015) No. 62/203,799.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Georgia Tech/Ajeet Rohatgi	ajeet.rohatgi@ece.gatech.edu	Task 1, FY14-15: Ion implantation to produce 156 mm B-emitters and PERT structures on n-Cz Si wafers for passivation/full size passivated back contact at NREL. Also, screen printed grids for NREL-passivated PERT cells.	95
Lightdrop Harvest LLC/Daniel Meier	dmeier@lightdropharvest.com	Task1, FY13-14: consult on Si cell process set up/development at NREL.	7.4
Colorado School of Mines/Sumit Agarwal	sagarwal@mines.edu	Task 1 and 2: PhD students working at NREL on Si cell projects: passivation, emitter, NiSi/Cu grid development, GaP on Si cell (FY14-15).	72
Arizona University/John Kouvetakis	jkouvetakis@asu.edu	Task 2, FY13-14: Synthesis of epitaxial Si-(III-V) and Si-(II-VI) compounds lattice-matched to Si.	112

CdTe Technology: Understanding the Defects

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Wyatt Metzger (Primary Investigator), 303-384-7939, Wyatt.Metzger@nrel.gov

Budget (FY13-FY15): $8.7M Agreement #: 25778

1. Agreement Description and Motivation

Current CdTe solar cell research is targeting 24% efficiency to drive module cost to less than $0.30/W, displace silicon market share, and reach grid parity. Recent efficiency gains have come largely by maximizing photocurrent. There is headroom to further improve efficiency by increasing open-circuit voltage (V_{oc}) and fill factor (FF), but record efficiency CdTe cells have been limited to V_{oc} less than 880 mV for the past two decades, whereas GaAs can attain V_{oc} of 1.10 V with a slightly smaller bandgap [2,3]. To overcome this barrier, we seek to understand and increase CdTe carrier lifetime and concentration, which have been limited to several nanoseconds and low 10^{14} cm^{-3} hole density.

2. Significant Achievements

The project succeeded in understanding and improving lifetime and doping in CdTe solar cells. We constructed a molecular beam epitaxy capability at NREL, grew epitaxial films and single crystal boules, and succeeded in growing polycrystalline films with grain sizes ranging from hundreds of nanometers to millimeters. Through novel characterization, we were able to quantify grain boundary recombination by type, and quantify and improve surface recombination. A combination of theory and experiments demonstrated that Te_{Cd} antisites and Cd vacancies are point defects that can increase recombination. By shifting from cation to anion doping, we were able to shift to a Cd-rich stoichiometry, reduce the number of these defects, place dopants on the Te sites, and establish simultaneously improved hole density, lifetime, and stability. In fact, we achieved radiatively limited (defect free) lifetimes over the range of 10^{16} cm^{-3} to low 10^{17} cm^{-3} in bulk CdTe. This doping range is ideal for solar cell absorbers, and the combination of long lifetime and hole density is competitive with state of the art III-V materials. Working with these materials, we were able to understand and overcome the photovoltage barrier that has limited CdTe technology for 50 years and make CdTe solar cells with world-record V_{oc} > 1V. The work has led to many records of inventions and patent applications, and created collaborative efforts with industry.

3. Modifications and Remaining Challenges

Our next three-year cycle is designed to build on the work from this period. This includes further establishing the science of doping with group V dopants in polycrystalline CdTe films. We will also work on reducing the role of grain boundary recombination, and forming ideal interfaces and contacts around these fundamentally different defect chemistries to enable next generation CdTe efficiencies at costs below $0.30/W.

4. FY13-FY15 Publications

Publications can be found on the www.nrel.gov/publications database.

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- Eric Colegrove, David S. Albin, Harvey Guthrey, Steve Harvey, James Burst, Helio Moutinho, Stuart Farrell, Mowafak Al-Jassim, Wyatt K. Metzger. "Phosphorus doping of polycrystalline CdTe by diffusion." 3396 42nd IEEE Photovoltaics Specialist Conference, New Orleans, LA June 14-19, 2015.
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5. FY13-FY15 Special Recognitions, Awards, and Patents

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- M. Reese, J. Burst, J. Duenow, D. Albin, T. Barnes, W. Metzger. “CdTe material for high lifetime and carrier concentration." Jan 2014 ROI Assigned" ROI-14-26.
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- Member Elected to Board of Directors of the AVS (October 2013). Term will be Jan. 2014-Dec. 2016. American Vacuum Society.
- Materials Research Society Fellowship (Suhuai Wei). 3/5/14
- NREL Research Fellow (Suhuai Wei). National Renewable Energy Laboratory

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
University of Illinois at Chicago	siva@uic.edu	MBE growth of CdTe solar cells	500

CIGS Thin-Film Technology: Overcoming Barriers to Increase Efficiency and Reduce Cost

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Miguel Contreras (Primary Investigator), 303-384-6478, miguel.contreras@nrel.gov

Budget (FY13-FY15): $7.831M Agreement #: 25779

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1. Agreement Description and Motivation

To lower the cost of $Cu(In,Ga)Se_2$ (CIGS) PV technology, the project aims at increasing efficiency and improving processes and materials used in the synthesis of CIGS PV materials. The efficiency improvement studies have a two-prong approach carried out by:

- Task 1 tackling improvements to photovoltage via the development of wide-gap CIGS materials
- Task 2 tackling improvements to photocurrent via wide-gap buffer layers.

Improvements to CIGS fabrication processes are covered under Task 3 focusing on improvements to the "2-step process" (the industrial process involving the selenization of metal precursors layers).

The final project milestone is the demonstration of a 22% CIGS solar cell.

2. Significant Achievements

- The project advanced the scientific understanding of the physics and materials science related to surface and interfaces of CIGS materials and PV devices. New physics insights and knowledge were attained.
- Developed new Hg-Probe technique to enhance our understanding of recombination mechanisms.
- Developed new Selective Photon TRPL technique to probe different depths in CIGS and elucidate new minority carrier physics.
- Advanced the efficiency of wide-gap CIGS materials beyond the state-of-the-art that existed at the start of this project. Demonstrated efficiencies above 18% for wide gap solar cell with V_{oc} above 800 mV.
- Best 1-sun result was a world-record efficiency of 20.7%.
- Best low-concentration efficiency result was a world-record 23.3%.
- Process development and optimization of the CBD ZnOS buffer layer resulted in the achievement of a 19.2% efficiency Cd-free solar cell with a 78.6% fill factor.
- KPFM measurements and absorber composition optimization demonstrated that ZnOS/CIGS solar cells are more sensitive to the absorber surface than CdS/CIGS because the ZnOS/CIGS junction is most likely a heterojunction compared to the CdS/CIGS buried homojunction.
- XPS measurements verified that ZnOS/CIGS devices have a much larger conduction band offset than CdS. Device physics modeling verified that ZnOS/CIGS charge transport across this large barrier is dominated by tunneling.
- Combining thin CdS with ZnOS unites the benefits of the optimal junction-doping of CdS and the larger bandgap of ZnOS.
- The NREL-developed CBD ZnOS buffer process was transferred to industry (US-based Stion), which resulted in a 13.5% efficiency 147W Cd-free solar panel manufactured on the pilot production line.
- Achieved 18.6% efficiency CIGS devices made from a two-step selenization process with a selenization time of 20 minutes.
- Increased the V_{oc} (by 48 to 70 mV) and efficiency of devices (by 1.5% to 1.9% absolute) by adding a KF post-deposition treatment to two-step processed CIGS.
- Detected a band-to-defect emission peak in CIGS by photoluminescence - the first report of this kind.
- Provisional patent application filed on the dopant-induced recrystallization (DIR) process that includes Sb in a CIGS precursor. DIR shows promise for lower-temperature processing of high-efficiency CIGS.

3. Modifications and Remaining Challenges

- The final milestone (22% efficiency) was missed, yet the project led to advancements in the state-of-the-art in CIGS technology. Best results we obtained were a world-record efficiency of 20.7% and efficiencies above 18% for wide gap solar cell with V_{oc} above 800 mV.
- There were many challenges to increase the efficiency of wide-gap CIGS solar cells. We discovered some key limitations to efficiency. Prioritizing areas of research and implementing solutions to overcome barriers was challenging.
- Development of the CBD ZnOS buffer layer required keen attention to both optimum device performance and performance variability. Variability and safety concerns led to the modification of the CBD process from zinc sulfate to zinc acetate due to the elimination of DMSO in the zinc acetate recipe.
- While the best CdS/CIGS solar cells incorporated alkali post-deposition treatment (PDT), strong electronic barriers prevented high efficiency ZnOS devices on PDT-absorbers. Further analysis and optimization are necessary to overcome this obstacle.
- The ZnOS/CIGS device structure has different reflection than the CdS/CIGS device structure, and more exploration is needed to find an optimal anti-reflection strategy for high-efficiency ZnOS solar cells.
- The rapid two-step selenization task was reviewed just before year 3. It was determined that the milestones focusing on time and efficiency were not as important as originally thought. The task was redirected to work on novel, Sb-assisted processing.
- Many questions about the new dopant-induced recrystallization (DIR) process remain unanswered.

4. FY13-FY15 Publications

Journal Publications

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- *Defect Segregation at Grain Boundary and its Impact on Photovoltaic Performance of CuInSe2* Yin, W. J.; Wu, Y.; Noufi, R.; Al-Jassim, M.; Yan, Y. Applied Physics Letters; Vol. 102 (19), 13 May 2013.
- *Electrical Conduction Channel Along the Grain Boundaries of Cu(In,Ga)Se2 Thin Films,* Jiang, C. S.; Repins, I. L.; Mansfield, L. M.; Contreras, M. A.; Moutinho, H. R.; Ramanathan, K.; Noufi, R.; Al-Jassim, M. M., Applied Physics Letters.
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- *Reformulation of Solar Cell Physics to Facilitate Experimental Separation of Recombination Pathways* Grover, S.; Li, J. V.; Young, D. L.; Stradins, P.; Branz, H. M., Applied Physics Letters; Vol. 103 (9), 26 August 2013.
- *Recombination Analysis of Cu(In,Ga)Se2 Solar Cells with Low and High Ga Compositions,* Li, J. V.; Grover, S.; Contreras, M. A.; Ramanathan, K.; Kuciauskas, D.; Noufi, R., Solar Energy Materials and Solar Cells; Vol. 124 May 2014.
- *Origin of reduced efficiency in Cu(In,Ga)Se2 solar cells with high Ga concentration: Structure separation vs. intrinsic defects,* B. Huang, S. Chen, H. Deng, L.-W. Wang, M. A. Contreras, R. Noufi, and S.-H. Wei, IEEE J. Photovoltaics 4, 477 (2014).
- *Direct imaging of enhanced current collection on grain boundaries of Cu(In,Ga)Se2 solar*

cells, J. Kim, S. Kim, C.-S. Jiang, K. Ramanathan, and M.M. Al-Jassim, Applied Physics Letters; Vol. 104 (6), 10 February 2014.

- *Defect properties of Sb- and Bi-doped CuInSe2: The effect of the deep lone-pair s states,* J.-S. Park, K. Ramanathan, and S.-H. WeiApplied Physics Letters; Vol. 105 (24), 15 December 2014.
- *Enhanced Performance in Cu(In,Ga)Se2 Solar Cells Fabricated by the Two-Step Selenization Process With a Potassium Fluoride Postdeposition Treatment,* L. M. Mansfield, R. Noufi, C. P. Muzzillo, C. DeHart, K. Bowers, B. To, J. W. Pankow, R. C. Reedy, and K. Ramanathan, IEEE Journal of Photovoltaics; Vol. 4 (6), November 2014.
- *Investigation of micro-electrical properties of Cu2ZnSnSe4 thin films using scanning probe microscopy* C.-S. Jiang, I.L. Repins, C. Beall, H.R. Moutinho, K. Ramanathan, M.M. Al-Jassim, Solar Energy Materials and Solar Cells; Vol. 132 January 2015.
- *Nanometer-scale surface potential and resistance mapping of wide-bandgap Cu(In,Ga)Se2 thin films,* C.-S. Jiang, M. A. Contreras, L. M. Mansfield, H. R. Moutinho, B. Egaas, K. Ramanathan, and M. M. Al-Jassim. Applied Physics Letters; Vol. 106 (4), 26 January 2015.
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- *Reducing interface recombination for Cu(In,Ga)Se2 by atomic layer deposited buffer layers,* Adam Hultqvist, Jian V. Li, Darius Kuciauskas, Patricia Dippo, Miguel A. Contreras, Dean H. Levi and Stacey F. Bent, Applied Physics Letters; Vol. 107 (3), 20 July 2015.

Conference Publications

- *Comparative Local Nanometer-Scale Resistance Mapping on Cu(In,Ga)Se2, Cu2ZnSnSe4, and CdTe Thin Films* C.-S. Jiang, I.L. Repins, L.M. Mansfield, R.G. Dhere, H.R. Moutinho, K. Ramanathan, R. Noufi, and M.M. Al-Jassim 39th IEEE Photovoltaic Specialists Conference.
- *Rapid fabrication of Cu(In,Ga)Se2 thin films from Se-containing precursors by the two-step selenization process* Lorelle M. Mansfield, Shogo Ishizuka, Clay DeHart, Marty Scott, Bobby To, Matthew R. Young, and Rommel Noufi 39th IEEE Photovoltaic Specialists Conference.
- *The Effect of Ga Content on the Recombination Behavior of Grain Boundaries in Cu(In,Ga)Se2 Solar Cells* Harvey Guthrey1, Miguel Contreras1, Mowafak Al-Jassim1, Spring 2014 MRS .
- *Electrical Characterization of Interface Recombination and its Dependence on Band Offset, Potential Barrier Height, and Inversion in Certain Heterojunction Solar Cells* Jian V. Li, Sachit Grover, Ingrid L. Repins, Brian M. Keyes, Miguel A. Contreras, Kannan Ramanathan, Rommel Noufi, Zhibo Zhao, and Feng Liao 40th IEEE PVSC.
- *Impact of buffer and absorber properties in the vicinity of the interface on wide-gap Cu(In,Ga)Se2 solar cell performance.* Ana Kanevce, Kannan Ramanathan, and Miguel Contreras. 40th IEEE PVSC.
- *Direct Evidence of a Cu(In,Ga)3Se5 Phase in a Bulk, High-Efficiency* Adam Stokes, Brian Gorman, Dave Diercks, Brian Egaas, and Mowafak Al-Jassim. 40th IEEE PVSC.
- *Nanoscale Electrical Properties of Wide-Bandgap Cu(In,Ga)Se2 and Cu2ZnSn(SSe)4 Thin Films* C. S. Jiang, M. A. Contreras, I. L. Repins, L. M. Mansfield, C. Beall, K. Ramanathan, and M. M. Al-Jassim 40th IEEE PVSC.
- *Band alignment of CBD deposited Zn(O,S)/Cu(In1-xGax)Se2 interface* Joel W. Pankow, K. Xerxes Steirer, Lorelle M. Mansfield, Rebekah L. Garris, Kannan Ramanathan, Glenn R. Teeter 40th IEEE PVSC.
- *The Effect of Ga Content on the Selenization of Co-evaporated CuGa/In Films and their Photovoltaic Performance* Christopher P.

Muzzillo, Lorelle M. Mansfield, Clay DeHart, Karen Bowers, Robert C. Reedy, Bobby To, Rommel Noufi, Kannan Ramanathan, and Timothy J. Anderson. 40th IEEE PVSC.

- *Direct Observation of Grain Boundary PN Junction Potentials in CIGS Using Photoemission and Low Energy Electron Microscopy (PELEEM)* Calvin K. Chan, Taisuke Ohta, Gary L. Kellogg, Douglas Pernik, Brian Korgel, Lorelle M. Mansfield, Rommel Noufi, and Kannan Ramanathan. 40th IEEE PVSC.
- *Efficient and Stable CIGS Solar Cells with ZnOS Buffer Layer* Rebekah L. Garris, Jian V. Li, Miguel A. Contreras, Kannan Ramanathan, Lorelle M. Mansfield, Brian Egaas, and Ana Kanevce. 40th IEEE PVSC.
- *Understanding Thin-Film PV Through Correlative Microscopy* H. Guthrie. MRS Spring 2015, Symposium B—Thin-Film Compound Semiconductor Photovoltaics.
- *Timr-Resolved PL Spectroscopy for Cu(In, Ga)Se2 Polycrystalline Films and PV Device;* D Kuciauskas, M. Contreras, B. Egaas, J. V. Li, H. Guthrey, P. Dippo, J. Pankow, and K. Ramanathan MRS Spring 2015, Symposium B—Thin-Film Semiconductor Photovoltaics.
- *Studying Defects and Interfaces in CIGS with High Resolution Correlative Microscopy* Jeffery A Aguiar, Harvey Guthrey, Adam Stokes, Toshihiro Aoki, Lorelle M Mansfield, Brian Eggas, Kannan Ramanathan, Mowafak Al-Jassim MRS Spring 2015, Symposium B—Thin-Film Compound Semiconductor Photovoltaics- B3.21.
- *Process Oriented Approach to CIGS Module Reliability* K. Ramanathan, L. Mansfield, R. Garris, T. Silverman and C, Deline PV Module Reliability Workshop.
- *Differences between CuGa/In and Cu/Ga/In Films for Selenization* Christopher P. Muzzillo, Lorelle M. Mansfield, Clay DeHart, Karen Bowers, Robert C. Reedy, Bobby To, Kannan Ramanathan, Timothy J. Anderson 42nd IEEE Photovoltaic Specialists Conference.
- *Development of Scanning Capacitance Spectroscopy on CIGS solar cells* C. Xiao, H.R. Moutinho, C.-S. Jiang, Y. Yan, D, Levi, and M.M. Al-Jassim 42th IEEE Photovoltaic Specialists Conference.
- *Optoelectronic investigation of Sb-doped Cu(In,Ga)Se2* Lorelle M. Mansfield, Darius Kuciauskas, Patricia Dippo, Jian V. Li, Karen Bowers, Bobby To, Clay DeHart, and Kannan Ramanathan 42nd IEEE Photovoltaic Specialists Conference.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- Suhuai Wei was selected as MRS Fellow in 2014.
- Adam Stokes, Best Student Presentation at IEEE PVSC 2014

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
University of Toledo, OH/ Yanfa Yan	University of Toledo/ Yanfa.Yan@utoledo.edu	Electron Microscopy Study of Photovoltaic Materials	23.4
Colorado School of Mines/ Brian Gorman	Colorado School of Mines/ bgorman@mines.edu	Investigation of the Structural Chemical and Electrical properties of Grain Boundaries in CIGS solar cells	40.0

Next-Generation Earth-Abundant Thin Film CZTS Photovoltaics

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Ingrid Repins (Primary Investigator), 303-384-7678, ingrid.repins@nrel.gov

Budget (FY13-FY15): $6.3M Agreement #: 25782

1. Agreement Description and Motivation

Kesterites (i.e., "CZTS," Cu2ZnSnSxSe4-x, and related alloys) can potentially function as an earth-abundant, polycrystalline, thin-film solar absorber. Such a material is expected to achieve the stability, low processing costs, low capital expenditure, and long lifetime associated with polycrystalline thin films, while maintaining very low materials costs - due to abundance - as manufacturing volumes approach TW levels. However, standards for device performance are high for any photovoltaic material: It is estimated that small-area device performance must exceed 20% to be a viable candidate for low module cost and resulting TW-scale production.
At the beginning of this program, world-record CZTS device efficiency was 10.1%, far from the >20% goal. Thus, in this program, we have focused on understanding and mitigating the detrimental defects in the kesterite system, to increase efficiency. We have developed fundamental understanding of defects in the kesterite materials system and made state-of-the-art kesterite devices. We have achieved this understanding by studying defects and interfaces in a variety of systems, ranging from theoretical models to simplified experimental samples and geometries to device-quality polycrystalline (PX) thin films.

2. Significant Achievements

Several significant achievements were made of the course of the research. These achievements are documented in publications (section 4), and several are highlighted in the list below:

- Created the first high-efficiency (>9%) vacuum-deposited kesterite device, leading to emergence of vacuum-processing of kesterites around the world.
- Synthesized novel kesterite alloys (e.g. $Cu_2ZnGeSe_4$, $Cu_2ZnSnSe_4$) and documented their structural and opto-electronic properties.
- Demonstrated agreement between first-principles and experimental band gap for Cu_2SnSe_3, correcting a long-standing misconception in the field.
- Measured band alignment and band-bending near the absorber-buffer interface and modeled the resulting effects on device performance.
- Created device model based on measured inputs that specifies how much voltage loss can be due to bulk versus other recombination mechanisms.
- Grew epitaxial CZTS on multiple substrates
- Developed *in operando* XPS measurement technique; characterized the effects of incident X-rays on photovoltage during XPS measurements.
- Performed comprehensive calculations of defect properties, including formation energies, energy level within gap, and tendency to interact with other defects. Experimentally verified properties for some defects.
- Demonstrated a general method for controlling metastable defect populations in devices via voltage-biased annealing.
- Demonstrated control of Cu-Zn disorder by device anneals, and correlated degree of disorder with device performance.
- Developed novel methods for *in situ* characterization of thin-film defect properties; quantified diffusion rates on CZT(S,Se) anion and cation sublattices.

3. Modifications and Remaining Challenges

The most significant challenge remaining for kesterite photovoltaics is achieving the required high performance (>20% power conversion efficiency) despite low defect formation energies.

Over the course of this research, the fundamental origin of these defect formation energies have been calculated, and their effects on the device both at the interface and in the bulk have been measured. We have been unable to reduce defect concentrations to levels that produce the desired device performance, and thus have missed a stretch quarterly progress indicator (Q4, FY15) involving demonstrating a 15%-efficient kesterite device. At present, the world record for kesterite devices is 12.6%, with all the performance improvement over the three-year period coming from improved current collection, rather than the more defect-sensitive "V_{oc} loss" (band gap minus open circuit voltage).

In the course of the work, minor modifications in the research direction were made to best address the observed efficiency losses. The original research proposed studies of defects in the bulk, of model systems (including fundamental calculations, and epitaxial growth), and of interfaces. When experimental results favorable properties at the grain boundaries of polycrystalline films, and epitaxial films included anti-phase domains, epitaxy was de-emphasized in favor of the other research thrusts.

4. FY13-FY15 Publications

Publications are listed below in chronological order. Manuscripts currently in progress or under review are not listed. Links to OSTI are included when available.

- H.R. Liu, S.Chen, Y.T. Zhai, H.J. Xiang, X.G. Gong, S.H. Wei, First-principles study on the effective masses of zinc-blend-derived Cu_2Zn-IV-VI_4 (IV = Sn, Ge, Si and VI = S, Se), Journal of Applied Physics, 112, 9, 93717, 2012.
- I.L Repins, M.J. Romero, J.V. Li, S.H. Wei, D. Kuciauskas, C.S. Jiang, C. Beall, C. DeHart, J. Mann, W.C. Hsu, G. Teeter, A. Goodrich, R. Nouf, Kesterite Successes, Ongoing Work, and Challenges: A Perspective From Vacuum Deposition, IEEE JOURNAL OF PHOTOVOLTAICS, 3, 1, 472-475, 2012.
- S. Chen, A. Walsh, X.G. Gong, S.H. Wei, Classification of Lattice Defects in the Kesterite Cu_2ZnSnS_4 and $Cu_2ZnSnSe_4$ Earth-Abundant Solar Cell Absorbers, ADVANCED MATERIALS, 25, 11,1522-1539, 2013.
- J.V. Li, D. Kuciauskas, M.R. Young, I.L. Repins, Effects of sodium incorporation in Co-evaporated $Cu_2ZnSnSe_4$ thin-film solar cells, APPLIED PHYSICS LETTERS, 102, 16,1 63905, 2013.
- Z. Wang, K.M Jones, A.G. Norman, J. Moseley, I.L. Repins, R. Noufi. Y. Yan, M.M. Al-Jassim, Electron microscopy study of individual grain boundaries in $Cu_2ZnSnSe_4$ thin films, IEEE PVSC 39, Tampa, FL, 2013.
- W.C. Hsu, I. Repins, C. Beall, C. DeHart, G. Teeter, B. To, Y. Yang, R. Noufi, The effect of Zn excess on kesterite solar cells, SOLAR ENERGY MATERIALS AND SOLAR CELLS,113,160-164, 2013.
- P. Xu, S. Chen, B. Huang, H.J. Xiang, X.G. Gong, S.H. Wei, Stability and electronic structure of Cu_2ZnSnS_4 surfaces: First-principles study, PHYSICAL REVIEW B, 88, 4,45427, 2013.
- I.L. Repins, H. Moutinho, S.G. Choi, A. Kanevce, D. Kuciauskas, P. Dippo, C.L. Beall, J. Carapella, C. DeHart, B. Huang, S.H. Wei, Indications of short minority-carrier lifetime in kesterite solar cells", JOURNAL OF APPLIED PHYSICS, 114, 8, 84507, 2013.
- W.C. Hsu, I. Repins, C. Beall, C. DeHart, B. To, W. Yang, Y. Yang, R. Noufi, Growth mechanisms of co-evaporated kesterite: a comparison of Cu-rich and Zn-rich composition paths, Progress in Photovoltaics: Research and Applications, 22, 1, 35-42, 2014.
- J.A. Polizzotti, I.L. Repins, R. Noufi, S.-H. Wei, D. Mitzi, The State and Future Prospects of Kesterite Photovoltaics ,Energy Environ. Sci., 6, 11, 3171, 2013.
- M. Bär, S. Pookpanratana, L. Weinhardt, R.G. Wilks, B. Schubert, B. Marsen, T. Unold, M. Blum, S. Krause, Y. Zhang, F. Khan, A. Ranasinghe, K. Ramanathan, I. Repins, M.A. Contreras, S. Nishiwaki, X. Liu, N.R. Paudel, M. Morkel, O. Fuchs, T.P. Niesen, W. Yang, F. Karg, E. Umbach, A.D. Compaan, W.N. Shafarman, R. Noufi, H.W. Schock, C. Heske, Soft x-rays shedding light on thin-film solar cell surfaces and interfaces, Journal of Electron Spectroscopy and Related Phenomena, 190, 47, 2013.
- P. Gecys, E. Markauskas, M. Gedvilas, G. Raciukaitis, I. Repins, C. Beall, Ultrashort pulsed laser induced material lift-off processing of CZTSe thin-film solar cells, Solar Energy, 102, 82, 2014.
- A. Kanevce, I. Repins, S.H. Wei, Impact of bulk properties and local secondary phases on the $Cu_2(Zn,Sn)Se_4$ solar cells open-circuit voltage, Solar Energy Materials and Solar Cells,133,119-125, 2015.
- H.R. Moutinho, M. Young, S. Harvey, C.-S. Jiang, C. Perkins, S. Wilson, M.M. Al-Jassim,

I.L. Repins, G. Teeter, Epitaxial growth of CZTS on Si substrates investigated with electron backscattered diffraction, IEEE PVSC 40, Denver, CO, 2014.

- C.-S. Jiang, M.A. Contreras, I.L. Repins, L.M. Mansfield, C. Beall, R. Ramanathan, M.M. Al-Jassim, Nanoscale Electrical Properties of Wide Bandgap $Cu(In,Ga)Se_2$ and $Cu_2ZnSn(S,Se)_4$ Thin Films, IEEE PVSC 40, Denver, CO, 2014.
- K. X. Steirer, R. Garris, C. Beall, A. Kanevce, K. Ramanathan, I. Repins, G. Teeter, C.L. Perkins, Photoelectron spectroscopy and photovoltaic device study of $Cu_2ZnSnSe_4$ and ZnO_xS_{1-x} buffer layer interface, IEEE PVSC 40, Denver, CO, 2014.
- B. Bob, J.V. Li, C. Beall, J. Carapella, C. Dehart, Y. Yang, I.L. Repins, Junction Formation and Interface Studies in CZTSe Solar Cells, IEEE PVSC 40, Denver, CO, 2014.
- A.E. Caruso, D.S. Pruzan, V. Kosyak, A. Bhatia, E.A. Lund, C. Beall, I. Repins, M.A. Scarpulla, Temperature Dependence of Equivalent Circuit Parameters Used to Analyze Admittance Spectroscopy and Application to CZTSe Devices, IEEE PVSC 40, Denver, CO, 2014.
- S.P. Harvey, C. Perkins, M. Young, H. Moutinho, S. Wilson, G. Teeter, Heteroepitaxial Growth of CZTS, IEEE PVSC 40, Denver, CO, 2014.
- S.G. Choi, S.-Y. Hwang, J. Li, C. Persson, Y.D. Kim, S.-H. Wei, I.L. Repins, Temperature dependent band-gap energy for $Cu_2ZnSnSe_4$ determined by spectroscopic ellipsometry, Solar Energy Materials and Solar Cells,130, 375-379, 2014.
- C.S. Jiang, I.L. Repins, C. Beall, H.R. Moutinho, K. Ramanathan, M.M. Al-Jassim, Investigation of micro-electrical properties of $Cu_2ZnSnSe_4$ thin films using scanning probe microscopy, Solar Energy Materials and Solar Cells,132, 324-347, 2015.
- D. Kuciauskas, I. Repins, A. Kanevce, J.V. Li, P. Dippo, C.L. Beall, Time-Resolved Recombination Analysis in Kesterite Polycrystalline Thin Films and Photovoltaic Devices With One-Photon and Two-Photon Excitation, Solar Energy Materials and Solar Cells, 136, 100-105, 2015.
- S.P. Harvey, I. Repins, G. Teeter, Defect Chemistry and Chalcogen Diffusion in Thin-Film $Cu_2ZnSnSe_4$ (C–ZTSe) Materials, Journal of Applied Physics, 117, 74902.
- I.L. Repins, J.V. Li, A. Kanevce, C. Perkins, K.X. Steirer, J. Pankow, G. Teeter, D.Kuciauskas, C. Beall, C. Dehart, J. Carapella, B. Bob, J.S. Park, S.H. Wei, Effects of deposition termination on CZTSe device characteristics, Thin Solid films, 582, 184-187, 2015.
- C. Wang, S. Chen, J. H. Yang, L. Lang, H. J. Xiang, X.G. Gong, A. Walsh, S.H. Wei, Design of I_2-II-IV-VI_4 Semiconductors through Element-substitution: the Thermodynamic Stability Limit and Chemical Trend, Chemistry of Materials, 26, 3411-3417, 2014.
- S.G. Choi, J. Kang, J. Li, H. Haneef, N.J. Podraza, C. Beall, S.-H. Wei, S.T. Christensen, I.L. Repins, Optical function spectra and bandgap energy of Cu_2SnSe_3, Applied Physics Letters, 106, 43902, 2015.
- H. Du, F. Yan, M. Young, B. To, C.S. Jiang, P. Dippo, D. Kuciauskas, Z. Chi, E. A. Lund, C. Hancock, W. M. Hlaing Oo, M. A. Scarpulla, G. Teeter, Investigation of combinatorial co-evaporated thin film Cu_2ZnSnS_4. I. Temperature effect, crystalline phases, morphology, and photoluminescence, Journal of Applied Physics, 115,173502, 2014.
- E.A. Lund, H. Du, W. M. Hlaing Oo, G. Teeter, M. A. Scarpulla, Investigation of combinatorial co-evaporated thin film Cu_2ZnSnS_4 (II): Beneficial cation arrangement in Cu-rich growth, Journal of Applied Physics, 115, 173503, 2014.
- J.S. Park, J.H. Yang, A. Kanevce, S. Choi, I.L. Repins, S.H. Wei, Ordering-induced direct to indirect band gap transition in multication semiconductor compounds, Phys Rev. B, 91, 7, 75204, 2015.
- M.E. Erkan, V. Chawla, I.L. Repins, M.A. Scarpulla, Interplay between Surface Preparation and Device Performance in CZTSSe Solar Cells: Effects of KCN and NH4OH Etching," Solar Energy Materials and Solar Cells, 136, 78-85, 2015.
- I.L. Repins, B. Egaas, L. Mansfield, M. Conteras, C. Muzillo, C. Beall, S. Glynn, J. Carapella, D. Kuciauskas, Fiber-Fed Time-Resolved Photoluminescence for Reduced Process Feedback Time on Thin Film Photovoltaics, Review of Scientific Instruments, 86, 103907, 2015. http://www.osti.gov/scitech/biblio/1190914
- D.S. Pruzan, A.E. Caruso, Y. Liu, I. Repins, C. Beall, M.F. Toney, M.A. Scarpulla, Sub-100 nm resolution 3-D Tomography of CZTSe

Using Transmission x-ray Microscopy, IEEE PVSC 42, New Orleans, LA, 2015.

- K. X. Steirer, R.L. Garris, J.V. Li, M.J. Dzara, P.F. Ndione, K. Ramanathan, I. Repins, G. Teeter, C.L. Perkins, Co-solvent Enhanced Zinc Oxysulfide Buffer Layers in Kesterite Copper Zinc Tin Selenide Solar Cells, Physical Chemistry Chemical Physics, early view on-line, 2015.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- Su-Huai Wei, **2014 MRS Fellow**, Materials Research Society, 3/7/2014.
- I.L. Repins, D. Kuciauskas, "Photoluminescence-based quality control for thin film absorber layers of photovoltaic devices," **U.S. patent #9,075,012**, 7/7/2015. (Cross-cutting work with CIGS, electro-optical characterization, and Bayh-Dole funds.)
- Repins, A. Kanevce, C.S. Jiang, Z.W. Wang, H. Moutinho, J.V. Li, C. Perkins, J.S. Park, S.-H. Wei, "Can CZTS Match the Performance of CIGS?," **Invited oral presentation**, 10th Workshop on the Future Direction of Photovoltaics," Tokyo, Japan 3/6/14.
- I.L. Repins, J.V. Li, A. Kanevce, C. Perkins, K.X. Steirer, J. Pankow, G. Teeter, D. Kuciauskas, C. Beall, C. Dehart, J. Carapella, B. Bob, J.S. Park, S.H. Wei, "Effects of deposition termination on CZTSe device characteristics," **invited oral presentation**, E-MRS Meeting, Lille, France, 5/28/14.
- S.G. Choi, "Optical properties of $Cu_2ZnSnSe_4$ and related compounds for thin-film photovoltaics," **invited oral presentation**, 3rd Korea-France Joint Symposium, Seoul, Korea 6/24/2014.
- H.R. Moutinho, R.G. Dhere, J. Duenow, C.S. Jiang, M. Young, S. Harvey, J. Moseley, D.L. Young, K. Alberi, M.M. Al-Jassim, I.L. Repins, G. Teeter, and W.K. Metzger, "Application of Electron Backscatter Diffraction in the Study of Photovoltaic Materials," **Invited oral presentation**, EBSD 2014 Pittsburgh, PA, 6/17/14.
- S.H. Wei, "First-principles design of multinary compounds for energy applications," **invited plenary oral**, International Conference on Ternary and Multinary Compounds, Niigata, Japan, 9/7/14.
- G. Teeter, S. Harvey, C. L. Perkins, C. Beall, S. Glynn, C. DeHart, K. Ramanathan, I. L. Repins, "Comparative Operando XPS Studies of CZTSe and CIGS Devices," **Invited Oral Presentation**, SunShot Initiative 3rd Thin-Film Photovoltaic Workshop, Newark, DE, 10/14.
- S. Choi, Current Status and Issues in Photovoltaic Technologies, **invited plenary oral**, UKC 2015, Atlanta, GA, 7/30/2015.

6. University and Industry Partners

The table below lists both funded and informal collaborations that occurred during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Stanford Linear Accelerator, Mike Toney	mftoney@slac.stanford.edu	Measure resonant x-ray diffraction on CZTSe to determine amount of disorder	15
University of Utah, Mike Scarpulla	scarpulla@eng.utah.edu	Electrical and chemical studies of CZTSe films and devices	0
University of Toledo, Nik Podraza	nikolas.podraza@utoledo.edu	Infrared ellipsometry of Cu_2SnSe_3 secondary phase	0
Fudan University, Congcong Wang	xggong@fudan.edu.cn	First-principles calculations of defect properties	0
University of Oslo, Clas Persson	clas.persson@fys.uio.no	First-principles calculations of optical properties	0
Lithuanian Center for Physical Sciences and Technology, Edgaras Markauskas	edgaras.markauskas@ymail.com	Laser scribing kesterites	0
Helmholtz-Zentrum Berlin für Materialien und Energie, Marcus Bär	marcus.baer@helmholtz-berlin.de	Advanced surface and interface characterization	0

Three-Year Photovoltaics (PV) Capital Reserve

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Greg Wilson, 303-384-7950, gregory.wilson@nrel.gov

Sarah Kurtz, 303-384-6475, sarah.kurtz@nrel.gov

Budget (FY13-FY15): $4.5M Agreement #: 15841

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1. Agreement Description and Motivation

The PV Capital Reserve was established in FY13 as part of the FY13-15 Lab Proposal Development Process by reserving $1.5 million of PV program funds each year for urgent capital needs within the National Center for Photovoltaics (NCPV).

2. Significant Achievements

During FY13-15, the Capital Reserve has provided funds to secure equipment for several PV research areas, including high efficiency cSi and Si tandems, measurement and characterization, and CIGS. It has also helped support the transition to a new process development and integration (PDIL) cleanroom.

Major equipment purchased with the Capital Reserve included:

- Horizontal furnace stack ($885K) and wafer cleaner ($1.48M) for the PDIL cleanroom
- Silicon reaction chamber for the III-V MBE cluster ($285K)
- Silicon atomic layer deposition tool ($180K)
- Ion beam miller ($189K)
- Solar simulator and uniformity mapper for CIGS research ($27K)
- Cryostat for low-temperature photoluminescence mapping ($81K).

Major cleanroom project related expenditures included:

- Electrical work related to PDIL tool relocations ($302k)
- Relocation of the PL imaging lab ($16k).

National Center for Photovoltaics Director's Initiative

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Greg Wilson, 303-384-7950, gregory.wilson@nrel.gov

Sarah Kurtz, 303-384-6475, sarah.kurtz@nrel.gov

Budget (FY13-FY15): $718K Agreement #: 25786

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1. Agreement Description and Motivation

The National Center for Photovoltaics (NCPV) Director's Initiative was established in the FY12 AOP to provide the NCPV Director with discretionary funds to use for emergency needs and, where possible, to seed new PV research activities. This practice was continued for FY13-15.

2. Significant Achievements

For the 3 years of this agreement, the NCPV Director's Initiative proved to be a critical source of funds for strategic and other unplanned expenses each year.

Funds were used for:

- Perovskite seed funding ($44K)
- FY15 perovskite agreement ($850K)
- HVPE reactor parts & assembly ($183K)
- FY13 severance expenses ($120K)
- Graduate student and postdoc support ($42K)
- Thin film and silicon workshops ($19K)
- Emergency equipment repairs ($8K)
- SunLamp responses outside of FY13-15 scope ($97K).

The funding for the FY15 perovskite agreement was moved into this agreement as carry-over from other PV agreements. The agreement was also used in FY14 to hold previously allocated funds for three NCPV strategic hires.

National Center for Photovoltaics (NCPV) Core Research and Development (R&D) Support

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Brian Keyes (Primary Investigator), 303-384-6695, brian.keyes@nrel.gov

Budget (FY13-FY16): $11.56M Agreement #: 26191

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1. Agreement Description and Motivation

The National Center for Photovoltaics (NCPV) Core Research and Development (R&D) Support Agreement provided the crosscutting infrastructure and support activities required by the core R&D agreements (CdTe, Silicon, Transparent Conducting Oxides (TCOs), CuInGaSe2 (CIGS), Organic PV (OPV), New Technique Development, CuZnSnS (CZTS), One-Sun III-V, Cell & Module Performance, III-V Multijunction, and Rapid Development of PV). The crosscutting support activities included:

- Task 1: Engineering Support: Design, Installation, Alteration and Maintenance of NCPV Capital Assets;
- Task 2: Informatics Support: Design and application of Data Handling & Analysis systems;
- Task 3: Measurements and Characterization (M&C) Support: Maintenance and Enhancement of M&C Capabilities;
- Task 4: Device Processing Support: Maintenance and Enhancement of Device Processing Capabilities.

The specific infrastructure activities were focused in individually distinct projects with the end result being 37 distinct project milestones throughout the course of the three-year effort. Of those 37 milestones, 32 were successfully completed during the three-year period, 2 will complete their readiness verification (RV) approval process within the first quarter of FY16, and 3 have been officially extended into FY16 to accommodate external construction delays associated with a new process development and integration (PDIL) cleanroom (see Section 3). The final project milestone is the demonstration of clean wafer transfer within the PDIL and the installation and connection to the scientific data network of research tools installed within the new cleanroom once construction is complete.

2. Significant Achievements

Engineering Support:

- Completed modifications to the Earth Abundant Deposition System enabling complete integration into the PDIL environment and controlled ambient transfer of samples between tools within that environment.
- Mitigated the principal contamination sources within the existing diffusion furnace facility resulting in improved performance and repeatability of performance-limiting Si device process steps.
- Completed construction of a III-V hydride vapor phase epitaxy (HVPE) deposition system enabling high impact research into the new and promising area of low-cost and high performance one-sun III-V technology development.
- Completed design, build and installation of a mapping stage for the Flash External Quantum Efficiency measurement system enabling increased throughput of combinatorial characterization and technology development efforts.
- Completed design, build and installation of an automated mapping stage for the current-voltage (I-V) measurement system enabling increased throughput of combinatorial characterization and technology development efforts.
- Designed and installed an automated gas handling system, including process control software, on the diffusion furnaces and demonstrated automated recipe-driven deposition that further improved process control and greatly improved project efficiency by freeing up researcher time.

- Completed upgrades to the W129 III-V metal organic chemical vapor deposition (MOCVD) reactor. This involved plumbing changes to add new sources, modernizing the controls for improved computer control, bringing the safety interlocks up to current standards, and writing control software to function similarly to the other MOCVD reactors in the group.
- Implemented upgrades to the HVPE system that increase system performance and repeatability via automated recipe driven control and improved control of dopant incorporation.
- Completed design and build of a novel environmental chamber allowing temperature and ambient control of a large number of samples with active bias during exposure and automated recipe-driven operation.
- Completed installation of a new epi-Si molecular beam epitaxy (MBE) chamber onto the manual cluster tool in the III-V lab enabling the study of lattice-mismatched structures.
- Developed a Customer Satisfaction Survey that was distributed yearly. The results were used to determine performance and improvement metrics.

Informatics Support:

- Designed and developed a generalized template for a deployable "data island" architecture that formed the basis of the Scientific Data Network (SDN). This approach has formed the basis for discussions across and beyond NREL concerning data handling and dissemination.
- Initiated data harvesting, set up data access, and aggregated data onto the SDN for a wide range of tools and laboratories including:
    - All PDIL M&C tools
    - All PDIL deposition tools
    - X-ray laboratory
    - Surface Analysis laboratory
    - Microscopy laboratory.

Measurements & Characterization Support:

- Completed automated data collection enhancements of the atomic force microscopy (AFM), photoluminescence (PL) and x-ray/ultraviolet photoelectron spectroscopy (XPS/UPS) instruments and demonstrated the ability to provide spatially resolved maps (44 predefined points across a 2in. x 2in. sample) of 1) root mean square (RMS) roughness, 2) PL peak position and 3) material properties required to determine interfacial band offsets: XPS core level intensity, XPS core level binding energy, XPS-derived composition or over-layer thickness, work function energy, and valence-band maximum energy. This greatly improved throughput of combinatorial research efforts by significantly reducing a measurement bottleneck in the process.
- Completed development of binary, ternary, and quaternary standards for quantitative compositional measurements using electron probe microanalysis (EPMA) and demonstrate compositional analysis using the new standards with an accuracy better than 1 atomic %.
- Completed development of a liquid nitrogen (LN2) cathodoluminescence (CL) system and demonstrated LN2-temperature CL images and spectra of CdTe, CIGS and CZTS. This increased the ability to perform faster low temperature surveys and reduced costs associated with previously required liquid helium measurements.
- Completed development of sample preparation procedures for electron backscattered diffraction (EBSD) examination and demonstrated high quality EBSD orientation maps of CdTe and CIGS with over 90% indexing capability. This unique measurement has now become a significant contributor to the polycrystalline research efforts at NREL.
- Completed development of electron-beam-induced current (EBIC)-guided transmission electron microscopy (TEM) sample preparation in the focused ion beam (FIB) tool and demonstrated the ability to acquire TEM images of targeted, site-specific areas in CdTe greatly improving the ability to correlate targeted microscopy with larger scale imaging data.
- Completed design and fabrication of a modification to the Rutherford backscattering spectrometry (RBS) tool enabling high-throughput data acquisition and analysis of 2"x2" combinatorial samples with 1mm spatial resolution and the ability to analyze compositional information from a system of non-overlapping RBS spectra (e.g. In-Zn-O material system).
- Extended the data acquisition (by incorporating spectral filtering and intensity modulation) and analysis methodology (by developing new simulation code) of the current-voltage measurement system. These improvements allowed for variability in excitation depth, increased signal-to-noise and

analysis routines that can now identify and quantify the dominant recombination pathway (e.g. interface, depletion region, quasi-neutral region and/or back contact recombination) in PV solar cells.
- Redesigned and rebuilt the liquid helium SEM cold stage and sample transfer system in order to extend the low temperature limit of the Cathodoluminescence (CL) capability to 6K, resulting in improved energy resolution and defect luminescence identification.
- Completed implementation of a software solution to track facility utilization. This calendar-based tool allows for scheduling tool usage and tracking that usage as a function of the supported project. The resulting usage data inform the discussions on tool prioritization, support, and resource allocation.

Device Processing Support:
- Completed development of new-to-NREL large area photolithography and e-beam metallization capabilities. This made the in-house capabilities compatible with the PDIL sample size.
- Develop a technique for transferring photomasks from acetate films created with a high resolution ink jet printer to glass plates and demonstrated that an acetate of a grid pattern for a 6” silicon solar cell can be transferred to an 8” iron oxide glass photomask while maintaining the same 10µm feature size as the original pattern on the acetate, +/- 2 µm. This laid the groundwork for a more cost effective and adaptable approach to in-house photomask development.
- Developed a large area Cu deposition capability and, building on the acetate-based mask development work, demonstrated the deposition of Ni/Cu contacts, with 10µm finger widths, and photolithography on 6” silicon wafers that will be metalized using a lift-off process (as opposed to the etch-back approach to removing unwanted material) in an e-beam vacuum deposition system.
- Improved NCPV expertise and understanding in high performance photoresist processes for front contact grid formations to solar cells, specifically with respect to the effect of humidity on the sidewall profiles of photoresists and gridlines and summarized the results via a white paper. The end result of this effort was the ability to process high aspect front contacts with an improved aspect ratio and sidewall profile, minimizing gridline-related losses.

3. Modifications and Remaining Challenges

- PDIL Cleanroom construction delays, external to this work effort, have delayed access to the PDIL Cleanroom. This has resulted in an inability to fully complete PDIL Cleanroom-related efforts. As of agreement end, March 2017,
    - the installation of the two new tools into the PDIL cleanroom (diffusion furnace and automated wet etch station) is nearly complete and the systems are scheduled to begin operation in Q3 FY17;
    - re-installation of existing tools into the PDIL cleanroom (Tystar RTA, Amerimade Wet Bench, MATECH Wave Etch) is nearly complete with readiness verification testing currently scheduled for completion in Q3 FY17;
    - The PDIL cleanroom tools (automated WetBench, Diffusion Furnace. Amerimade WetBench, and Optical Furnace) have been connected to the Research Data Network. Data harvesting capabilities are scheduled for implementation in Q3 FY17.

4. FY13-FY15 Publications

As a general rule, the NCPV Core R&D Support Agreement provided scientific infrastructure support and the resulting science and publications enabled by this support is covered within the corresponding research agreement reports. There are the few exceptions noted below.

- Jian V. Li, Sachit Grover, Ingrid L. Repins, Brian M. Keyes, Miguel A. Contreras, Kannan Ramanathan, Rommel Noufi, Zhibo Zhao, and Feng Liao, “Electrical Characterization of Interface Recombination and its Dependence on Band Offset, Potential Barrier Height, and Inversion in Certain Heterojunction Solar Cells” from IEEE PVSC, 2014 (10.1109/PVSC.2014.6925013)
- Robert R. White and Kristin Munch, “Handling Large and Complex Data in a Photovoltaic Research Institution”, from the Fall 2013 MRS Meeting (http://arxiv.org/abs/1403.2656)

5. FY13-FY15 Special Recognitions, Awards, and Patents

None

6. University and Industry Partners

The NCPV Core R&D Support Agreement did not collaborate with any external industrial or university partners.

Photovoltaic (PV) Partnering and Business Development

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Brian Keyes, 303-384-6695, brian.keyes@nrel.gov

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Sarah Kurtz, 303-384-6475, sarah.kurtz@nrel.gov

Paul Basore, 303-384-6420, paul.basore@nrel.gov

Budget (FY13-FY16): $8.99M Agreement #: 25784

1. Agreement Description and Motivation

The PV Partnering and Business Development Agreement's objective is to advance PV technology towards SunShot goals via collaborative partnerships, outreach activities, and techno-economic analysis. Key components of this agreement include:

- The Non-proprietary Partnering Opportunities (NPO) task that enables collaborative research between NREL and both companies and universities to advance and publish scientific understanding in critical areas of PV technology development. Partners were selected based upon a merit review of the proposed work. Selected partners gained access to NREL's PV capabilities that enable them to achieve their NPO research objectives. DOE funds supported NREL's contribution to these collaborative research projects.
- The Proprietary Partnering Opportunities (PPO) task aimed at growing the National Center for Photovoltaics (NCPV) portfolio of funds-in activities, both in the form of Technology Partnership Agreements (TPAs) and funds acquired through competitive solicitations (or Funding Opportunity Announcements – FOAs).
- The NCPV outreach task focused on workforce development opportunities for graduate students and postdocs. Two key activities associated with this task involved NCPV Fellowship program and the Hands-On PV Experience (HOPE) workshop.
- The Techno-economic analysis task aimed at training PV specialists to conduct techno-economic analysis that guides program direction.

The final project milestones include: documenting at least 30 peer-reviewed journal publications associated with the non-proprietary partnership opportunity (NPO) task; cost at least $2.8M in Technology Partnership Agreement (i.e. non-DOE) funded PV research in FY15; Conduct the Annual Hands On Photovoltaic Experience (HOPE) event; and conduct techno-economic analysis on III-V substrate reuse, Si tandems, and polycrystalline thin film tandems.

2. Significant Achievements

- The Non-proprietary Partnering Opportunity (NPO) program was established and eleven (11) selection rounds occurred from FY13 to FY15 that evaluated sixty-three (63) applications, resulting in twenty-nine (29) separate collaborative support projects. These projects involved 25 different principal investigators from 21 different institutions (15 universities and 6 private companies).
- As of September 2015, the NPO program has, and is, involved in numerous peer-reviewed journal articles: 14 published, 6 submitted, and 16 manuscripts in progress with more to come as the final projects are completed. Additionally, research results have been presented at major scientific conferences including the Spring MRS Meeting, the American Physical Society Conference and the IEEE Photovoltaic Specialists Conference, among others. A few notable scientific contributions include:
    - Development of a contactless inline characterization technique capable of

simultaneously determining bulk and surface recombination properties,
    - Quantified and identified ZnS as a preferred buffer layer for $Cu(In,Ga)Se_2$ devices,
    - Demonstrated rapid thermal processing (RTP) as a successful and low thermal budget approach to activating ZnTe:Cu-based back contact layers for CdTe solar cells,
    - Demonstrating Si δ-doping as an effective method to fill the subband levels of quantum dots with electrons and tune both the interband carrier transitions and carrier lifetimes towards improved intermediated band solar cells,
    - Optimization of CdTe solar cells via use of an oxygenated CdS window layer,
    - Demonstrated Ga_2O_3 as a suitable buffer layer for earth abundant Cu_2O-based solar cells,
    - Demonstrated in-situ photoluminescence as a potentially useful technique to monitor SiN_x wet etching for metal plating applications,
    - Guided earth abundant Cu_2O development by determining band offsets of n-type electron-selective contacts,
    - Provided a proof-of-concept framework for separating recombination processes in thin silicon wafers using transient free-carrier absorption spectroscopy
- Despite a difficult commercial PV business development environment, the NCPV was able to bring in over $7.1M in PV-related non-DOE funding between FY13 and FY15.
- FY15 AOP investments in business development (BD) have resulted in significant progress towards DoD funded activities. We spent a small fraction of these funds working with the Army Research Lab ($3,900) to develop new work on low cost III-Vs via HVPE and expect a substantial return on this investment in late FY16 or early FY17.
- FY15 AOP BD investments along with BD investments from NREL enabled the establishment of a major new commercial CRADA related to III-V multi-junctions for CPV applications.
- Each year, about a dozen graduate students were hosted at NREL for a week from major U.S. universities to learn about PV. The students benefited from the interaction with students and professors from the other universities and learned from NREL scientists many details about PV device fabrication, characterization, and application.
- Seven members of the NCPV staff, representing four different NREL Centers, directly engaged with three staff from NREL's Strategic Energy Analysis Center to learn the basic concepts and methods of techno-economic analysis. Weekly meetings and individual research effort led to the preparation of three papers suitable for publication.
- Key results of the techno-economic analysis include: (1) The cost of steps that are necessary to reuse a substrate for III-V epitaxial deposition exceed the substrate cost itself after only about twenty reuse cycles, for all three of the reuse methods investigated, and (2) Thin-film tandem structures have the potential to reduce LCOE under a limited range of circumstances, such as residential rooftops where BOS cost is high, and (3) The projected LCOE reduction using a III-V/Si tandem module in a utility-scale system meeting SunShot BOS cost goals is at most 15%, regardless of the III-V cell cost.

3. Modifications and Remaining Challenges

- The NPO program will complete the remaining three (3) collaborative support projects in FY16. NREL will provide a mid-year progress report and an Annual Milestone Report summarizing the FY13-16 NPO Program and documenting status of the milestone of publication of thirty (30) NPO peer-reviewed journal publications. While the interim milestone of 20 publications was missed, it is our belief that this is more a result of underestimating the time frame for publication than the potential and expectation for publication. The NPO Program currently has >30 manuscripts (accepted, submitted and in-progress), and is well on its way to achieving the final project milestone.
- FY15 saw a substantial increase in business development activities within the NCPV and more broadly, at NREL. Even though we missed our $2.8M funds-in milestone (actual was $2M), significant progress was made in developing a more sophisticated approach to business development and the following priorities have been established for FY16:
    - NREL will emphasize and target larger key accounts. Two of these accounts,

Abengoa and First Solar, will be managed by NCPV staff in FY16
  - We will rework the NREL web presence to better facilitate PV and related materials research business development .
  - The NCPV will continue to serve as NREL's PV and PV reliability focused institute that will coordinate business development efforts within the MCST Directorate. A committee consisting of the MCST Center Directors and the PV Program Manager will establish business development priorities monthly.
- The original scope of the techno-economic analysis task was to publish one paper on reuse of substrate wafers for III-V solar cells, and provide outlines for two additional papers on the economics of tandem cell structures. In July 2015, funds were reallocated within the project to support expansion of the two outlines into complete papers -- one for III-V on Si tandems, and one for thin-film on CIS tandems.

4. FY13-FY15 Publications

- Progress in Photovoltaics: "Optimization of CdTe thin-film solar cell efficiency using a sputtered, oxygenated CdS window layer" (http://199.171.202.195/doi/10.1002/pip.2578/epdf)
- Journal of Applied Physics: "Ellipsometric studies of Al x Ga1− x As0.5Sb0.5 (0.0 ≤ x ≤ 0.6) alloys lattice-matched to InP(100)" (http://dx.doi.org/10.1063/1.4861614)
- Advanced Materials: "Atomic Layer Deposited Gallium Oxide Buffer Layer Enables 1.2 V Open-Circuit Voltage in Cuprous Oxide Solar Cells" (http://dx.doi.org/10.1002/adma.201401054)
- Applied Physics Letters: "In-situ photoluminescence imaging for passivation-layer etching process control for photovoltaics" (http://dx.doi.org/10.1063/1.4891642)
- Applied Physics Letters: "Band offsets of n-type electron-selective contacts on cuprous oxide (Cu2O) for photovoltaics" (http://dx.doi.org/10.1063/1.4905180)
- Journal of Applied Physics: "Proof-of-concept framework to separate recombination processes in thin silicon wafers using transient free-carrier absorption spectroscopy": (http://dx.doi.org/10.1063/1.4914160)
- IEEE Journal of PV: "Simultaneous Measurement of Minority-Carrier Lifetime in Single-Crystal CdTe Using Three Transient Decay Techniques" (http://dx.doi.org/10.1109/JPHOTOV.2014.2339491)
- Journal of Applied Physics: "Dual-sensor technique for characterization of carrier lifetime decay transients in semiconductors" (http://dx.doi.org/10.1063/1.4903213)
- Journal of Applied Physics: "Ellipsometric characterization and density-functional theory analysis of anisotropic optical properties of single-crystal α-SnS" (http://dx.doi.org/10.1063/1.4886915)
- Journal of Crystal Growth: "Effect of silicon delta-doping density on optical properties of type-II InAs/GaAsSb quantum dots" (http://dx.doi.org/10.1016/j.jcrysgro.2014.08.009)
- Semiconductor Science and Technology: "Impact of delta-doping position on photoluminescence in type-II InAs/GaAsSb quantum dots" (http://dx.doi.org/10.1088/0268-1242/30/3/035006)
- Solar Energy Materials and Solar Cells: "Controlled activation of ZnTe:Cu contacted CdTe solar cells using rapid thermal processing" (http://dx.doi.org/10.1016/j.solmat.2014.10.045)
- Applied Physics Letters: "Reducing interface recombination for Cu(In,Ga)Se2 by atomic layer deposited buffer layer" (http://dx.doi.org/10.1063/1.4927096)
- IEEE Journal of PV : "Photovoltaic Material Characterization with Steady-State and Transient Photoluminescence" (http://dx.doi.org/10.1109/JPHOTOV.2014.2361015)
- As of September 2015, there are also four (5) manuscripts submitted to peer-reviewed journals and fourteen (17) manuscripts in progress for eventual submission to peer-reviewed journals.
- One techno-economic analysis paper was submitted to *Progress in Photovoltaics*, two additional papers are currently undergoing internal review prior to submission to peer-reviewed journals.

5. FY13-FY15 Special Recognitions, Awards, and Patents

- None to Report

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Table 1: Non-proprietary Partnering Opportunity Collaborators. The funding values are *estimates* of the NREL funds associated with each project.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity
Colorado State University / W.S. Sampath	sampath@engr.cpolostate.edu	Measurement and Characterization for Higher Efficiency CdTe Solar Cells
Oregon State University / Gregory Herman	greg.herman@oregonstate.edu	Characterization of Films and Interfaces Fabricated From of Earth Abundant Nanomaterials for Low Cost Photovoltaics
MicroLink Devices / Rao Tatavarti	rtatavarti@mldevice.com	Evaluation of Dislocation Density in ELO Metamorphic Buffer Layers on 6" GaAs Substrates
University of Utah & University of South Florida / Mike Scarpulla	scarpulla@eng.utah.edu	Understanding the Stoichiometry of Thin Film CdTe at the Surface and in the Bulk
University of California, Irvine / Matt Law	matt.law@uci.edu	Surface Analysis of Hole-Conducting Nanocrystalline Films for >20% Efficient Tandem Dye-Sensitized Solar Cells
MicroLink Devices / Jessica Adams	jadams@mldevices.com	Extensive Characterization of InAlAsSb for Wide Bandgap Subcell in Multi-junction Solar Cell Lattice-Matched to InP
Integrated Photovoltaics / Dirk Weiss	dirkweiss@gmail.com	Defect studies in kerfless c-Si wafer replacement
Massachusetts Institute of Technology / Tonio Buonassisi	buonassisi@mit.edu	Interface characterization of novel buffer-layer materials on photovoltaic absorbers such as Cu2O and SnS
Massachusetts Institute of Technology / Tonio Buonassisi	buonassisi@mit.edu	Decoupling bulk and surface-limited lifetimes in thin kerfless crystalline silicon
University of Delaware / William Shafarman	wns@udel.edu	Enhancement of Optical Simulation Software PV Optics to Include the Capacity to Model Thin Film Polycrystalline Solar Cell Device Structures
Lakewood Semiconductors / Richard Ahrenkiel	rahren@mac.com	Technique Development for the Simultaneous Measurement of Photoconductivity and Photoluminescence Decay in Photovoltaic Materials
Crystal Solar / TS Ravi	tsravi@xtalsolar.com	Analysis of Impurities and Defects in Epi-Si Solar Wafers and Cells
University of Minnesota / Stephen Campbell	campb001@umn.edu	Characterization of Thin Film Tunnel Junction Materials for Photovoltaics
Applied Materials / James Gee	James_Gee@amat.com	Single crystal silicon sawing: morphological, subsurface and minority carrier lifetime properties for diamond wire cutting of thin (100µm) 1-0-0 and 1-1-1 orientation wafers
Penn State University / Jeffrey Brownson	jrb52@psu.edu	Carrier and Band Properties in Tin Monosulfide Thin Films
Arizona State University / Christiana Honsberg	christiana.honsberg@asu.edu	Measurement of Physical Mechanism for Intermediate Band Solar Cells
Scifiniti / Dirk Weiss	dirkweiss@gmail.com	The Origin of Stress and Mitigation of Stress-Induced Defects in Scifinti's SmartWafer

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity
Arizona State University / Mariana Bertoni	bertoni@asu.edu	Defect Studies in Chalcogenides Solar Cells
Colorado School of Mines / Eric Toberer	etoberer@gmail.com	Control of Optoelectronic Properties of Zinc Tin Nitride Photovoltaic Absorbers
Colorado School of Mines / Colin Wolden	cwolden@mines.edu	Advanced Optoelectronic Characterization of CdTe Solar Cells Produced by Non-equilibrium Processing
Stanford University / Stacey Bent	sbent@stanford.edu	Investigation of the electronic properties of buffer layer/Cu(In,Ga)Se2 interfaces using one and two photon excitation photoluminescence
University of Utah / Mike Scarpulla	scarpulla@eng.utah.edu	Investigation of Grain Boundary Recombination and Band Structure in Cu2ZnSnS4 by Cathodoluminescence
Penn State University / Joan Redwing	jmr31@psu.edu	Silicon chalcogenides - earth abundant, silicon-compatible top cell materials
Binghamton University / Tara Dhakal	tdhakal@binghamton.edu	Study of Alternative Cd-free n-type Buffer Layers for use with Cu2ZnSnS4 (CZTS) Solar Cell Absorbers
Purdue University / Peter Bermel	pbermel@purdue.edu	Calculating Surface and Bulk Recombination in Thin-Film Indium Phosphide using Time-Resolved Photoluminescence and Photoluminescent Excitation Spectroscopy
University of North Carolina at Charlotte / Abasifreke Ebong	aebong1@uncc.edu	Development of low-cost screen-printable metal alternative to Ag for silicon solar cells
Purdue University / Rakesh Agrawal	agrawalr@purdue.edu	In Depth Characterization and Control of Potential Fluctuations for Increasing VOC in CZTS
University of New Orleans / Weilie Zhou	wzhou@uno.edu	Crystallographic and Stability Observation of Metal Halide Perovskite Solar Cells via Advanced Microscopy Techniques
Arizona State University / Mariana Bertoni	bertoni@asu.edu	Investigation of Molybdenum Oxide and Oxysulfides - Silicon Interfaces for High Efficiency PV Applications

Overcoming Spatial, Energy, and Temporal Limits in Characterization of Electronic, Optical, and Structural Properties of PV Materials

Performing Organization: National Renewable Energy Laboratory

Key Technical Contact: Dean Levi (Primary Investigator), 303-384-6605, dean.levi@nrel.gov

Budget (FY13-FY15): $3.2M Agreement #: 25777

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1. Agreement Description and Motivation

The goal of this project is to develop a complimentary set of highly advanced electronic and optical materials characterization techniques with sub-micron spatial resolution capable of clearly determining the electronic properties and physical origins of the defects and impurities that control doping and lifetime in thin-film PV materials. Limitations in doping and minority carrier lifetime are the two most recognized barriers to advances in efficiency in both industrial and emerging thin-film PV. This is largely because the knowledge base of electrically active defects in thin-film PV materials is significantly lacking. A major reason for this dearth of knowledge is the lack of characterization techniques capable of resolving the properties of impurities and defects that are inhomogeneously distributed between grains, grain boundaries, and interfaces on a sub-micron scale.

The project is divided into two major tasks; (1) development of nanoscale electrical characterization techniques, and (2) development of nanoscale optical characterization techniques. The nanoscale electrical characterization task is divided into three subtasks: (1) expanded energy range admittance spectroscopy / deep level transient spectroscopy (AS/DLTS); (2) scanning probe microscope (SPM)-based scanning capacitance spectroscopy (SCS); and (3) SPM-based AS/DLTS spectroscopy. The nanoscale optical characterization task is divided into two subtasks: (1) development of two photon excitation (2PE) photoluminescence (PL) and time-resolved photoluminescence (TRPL) microscopy with sub-micron resolution, and (2) development of tip-enhanced Raman spectroscopy (TERS) with sub-100nm resolution. The final project milestone consists of individual milestones for each of these five subtasks, as described in the following discussions.

2. Significant Achievements

Subtask 1.1 Expanded energy range AS/DLTS spectroscopy

- This subtask takes advantage of the latest development in state-of-the-art RF lock-in amplifier to expand the rate window of the system by 4 orders of magnitude. In addition, we developed thermal engineering approaches in a cryogenic probe station to expand the temperature window, especially at the low end. Ultimately, we built a unique AS/DLTS apparatus with wide rate window (10^{-2} to 10^{8} Hz) and wide temperature window (10-475 K) and succeeded in observing previously undetected ultra-shallow (<30 meV) and ultra-deep (>700 meV) defect levels in CIGS thin-film PV materials.

Subtask 1.2: SPM-based scanning capacitance spectroscopy

- Developed scanning spreading resistance microscopy (SSRM) for resistance mapping on plain-view thin film PV materials and on device cross-sections. Resolutions better than 40 nm on Si and 30 nm on CIGS were demonstrated.
- Applied SSRM to map the local resistivity on CIGS, CZTS, and CdTe thin films and devices. Conduction channels were observed on CIGS grain boundaries and on some CZTS grain boundaries, but not on CdTe films.
- Developed a procedure for the growth of a high-quality native oxide on cross sections of CIGS/CdS and CZTS/CdS solar cells that enables acquisition of reliable SCM and SCS data.

- Successfully analyzed cross sections of CIGS/CdS and CZTS/CdS solar cells by SCM and SCS, mapping the variation of the signal on the p- and n- sides of the junction, as well as in the depletion region.
- For the first time, provided conclusive evidence of a buried homojunction in CIGS/CdS solar cells. Correlations of the SCM/SCS data with AFM and SEM images provided the relative location between the electrical and metallurgical junctions with resolution better than 30 nm. The electrical junction was measured to be 40-45 nm from the CIGS/CdS metallurgical junction in an NREL CIGS solar cell.

Subtask 1.3 SPM-based AS/DLTS spectroscopy

- In a first-ever measurement we demonstrated high-sensitivity (~10 aF) capacitance measurements in a conductive AFM without the traditional resonant-circuit method employed by SCM. This was enabled through design and fabrication of custom IC's and specially designed high gain, high sensitivity pre-amplifiers. This enables absolute measurement of capacitance, which is required for microscopic AS/DLTS.
- We demonstrated a proof-of-concept setup for high spatial resolution, single-point operation of AS/DLTS measurement and succeeded in collecting data from Si, GaAs, and CIGS PV materials. Capacitance transient data in CIGS materials revealed the first-time microscopic observation of an electrostatic barrier to trap filling, which had been only observed on macroscopic scale previously and was thought to be responsible for the metastability phenomenon in CIGS.

Subtask 2.1: Two-photon excitation PL and TRPL with sub-micron spatial resolution

- Developed methods and hardware for nonlinear, sub-bandgap two-photon excitation time-resolved photoluminescence (2PE-TRPL) spectroscopy for analysis of minority carrier lifetime in the bulk of PV absorbers.
- Applied 2PE-TRPL to analyze the effect of intrinsic defects in CdTe on the photoexcited minority carrier lifetime. Determined that Cd-rich stoichiometry has lower non-radiative recombination. This result was published in Physical Review Letters and has contributed to significant advances in record high minority carrier lifetime in epitaxial and polycrystalline thin film CdTe materials and devices.
- Designed and constructed two nonlinear optical microscopes, one for 2PE-TRPL lifetime analysis and one for 2PE low-temperature PL spectroscopy down to 4 degrees Kelvin. Demonstrated 0.5-micron optical resolution in both microscopes. Both microscopes are capable of sectioning samples in (x,y) and (x,z) with 0.5 um lateral and 1.5 um axial resolution.
- Utilized the 2PE-TRPL microscope to measure the interface recombination velocity at the buried interface in a CdTe epitaxial sample. This is a unique and valuable result that is to our knowledge the first time this has been accomplished.
- Utilized a combination of analytical models and TCAD numerical simulations to develop advanced understanding of the role of carrier transport and how it limits effective spatial resolution in 2PE-TRPL microscopy. We are able to conclude that spatial resolution is determined by the optical resolution when the photoexcited carrier diffusion length is less than the laser spot diameter. When the diffusion length is larger than the laser spot diameter diffusion dominates the spatial resolution in 2PE-TRPL and 2PE-PL measurements. This presents the opportunity to derive carrier transport and lifetime information from a single 2PE-TRPL measurement, yet can limit spatial resolution in this embodiment of the 2PE-TRPL method.

Subtask 2.2: Development of Tip-Enhanced Raman Scattering (TERS) Spectroscopy

- Demonstrated the capability of Raman spectroscopy mapping to locate secondary phases in CIGS and CZTS thin film PV materials.
- Adapted atomic force microscopy (AFM)-based TERS technique to thin film PV materials. Demonstrated TERS measurements of $CuInGaSe_2$ thin film sample with the spatial resolution of <40 nm.

3. Modifications and Remaining Challenges

Subtask 1.2: SPM-based scanning capacitance spectroscopy

- For SSRM a further decrease of the contact resistance between probe and sample will allow for a quantitative and accurate mapping of the local resistivity.
- In SCS determining the location of the metallurgical junction using AFM and SEM is a

challenge because of the small roughness required for SCM/SCS analysis. Developing a more efficient process to for surface preparation will enable application of this method to more samples in less time.

- Modeling is a challenge because of the complex SCM/SCS signal on the depletion region. However, its development, and correlation with the experimental data will allow for better confirmation of the location of the electrical junction.

Subtask 1.3 SPM-based AS/DLTS spectroscopy

- In the original proposed work, we planned to use the AFM tip to directly contact the sample surface and form a Schottky contact. Due to thermal drift we ran into significant challenges in achieving reliable electrical contact between the AFM tip and the sample. Although we had some limited success using this method, we finally had to resort to using deposited metal contacts to form a high-quality Schottky contact to the sample. These metal contacts were reduced then to microscopic-scale areas (a few μm^2) meaningful for this subtask. When the AFM tip contacts these microscopic-scale metal areas, the electrical contact is then reliable. This modification of the original proposed work is not a fundamental issue but is outside the scope of the original project. For future applications we anticipate the use of high-resolution lithography for contact deposition will enable scanning-mode operation. The final milestone status of this subtask is 75% complete.

Subtask 2.1: Two-photon excitation PL and TRPL with sub-micron spatial resolution

- It became clear through this work that photoexcited carrier diffusion limits spatial resolution in high quality samples, with diffusion lengths of 5 – 10 microns or more. More sophisticated optical methods will be required to overcome the diffusion length limit to spatial resolution in 2PE-PL microscopy.

Subtask 2.2: Development of Tip-Enhanced Raman Scattering (TERS) Spectroscopy

- Application of TERS has been relatively successful in studies of highly conductive nanoscale materials such as graphene and carbon nanotubes. To accommodate low conductivity and rough surfaces typical of polycrystalline PV thin film materials, we developed TERS with atomic force microscopy (AFM) tips whereas the scanning tunneling microscopy (STM) tips are more widely used for TERS technique. However, a number of technical challenges remain:
  - Acquisition of high-performance reliable tips
  - Preparation of polycrystalline PV thin film samples for TERS measurement (cross-sectional scan in particular)
  - Precise alignment of laser beam onto the tip end in a reproducible manner

4. FY13-FY15 Publications

- J. V. Li, “DLTS study of defects in thin-film photovoltaic materials,” MRS Spring Meeting, 2014 (invited)
- C.-S. Jiang, M.A. Contreras, L.M. Mansfield, H.R. Moutinho, B. Egaas, K. Ramanathan, and M.M. Al-Jassim, Nanometer-scale potential and resistance mapping of wide-bandgap $Cu(In,Ga)Se_2$ thin films, Appl. Phys. Lett. 106, 43901 (2015).
- H. Li, C.-S. Jiang, W.K. Metzger, C.K. Shih, and M.M. Al-Jassim, Nanometer-resolution resistance mapping across the junction of CdTe solar cell, IEEE Journal of Photovoltaic, 5, 395 (2015).
- C. Xiao, H.R. Moutinho, C.-S. Jiang, B. To, D. Levi, Y. Yan, M.M. Al-Jassim, Development of scanning capacitance spectroscopy of CIGS solar cells, Proc. of the 42th IEEE PVSC (New Orleans, 2015), in press.
- C.-S. Jiang, I.L. Repins, C. Beall, H.R. Moutinho, K. Ramanathann and M.M. Al-Jassim, Investigation of micro-electrical properties of $Cu_2ZnSnSe_4$ thin films using scanning probe microscopy, Solar Energy Materials and Solar Cells, 132, 342 (2014).
- C.S. Jiang, Miguel A. Contreras, Ingrid L. Repins, Lorelle M. Mansfield, Carolyn Beall, Kannan Ramanathan, and Mowafak M. Al-jassim, Nanoscale electrical properties of wide bandgap $Cu(In,Ga)Se_2$ and $Cu_2ZnSnSe_4$ thin films, 40th IEEE PVSC, Denver, CO, 2014.
- H. Li, C.-S. Jiang, W. Metzger, C.-K. Shih, and M.M. Al-Jassim, Nanometer-scale study of resistance on CdTe solar cell devices, 40th IEEE PVSC, Denver, CO, 2014.
- C.-S. Jiang, H.R. Moutinho, R.G. Dhere, and M.M. Al-Jassim, The nanometer-resolution local electrical potential and resistance mapping of CdTe thin films, IEEE Journal of Photovoltaics, 3, 1383 (2013).

- C.-S. Jiang, I.L. Repins, L.M. Mansfield, R.G. Dhere, H.R. Moutinho, K. Ramanathan, R. Noufi, and M.M. Al-Jassim, Comparative Local Nanometer-Scale Resistance Mapping on Cu(In,Ga)Se2, $Cu_2ZnSnSe_4$, and CdTe Thin Films, 39th IEEE PVSC, Tampa, FL, 2013.
- C.-S. Jiang, I.L. Repins, L.M. Mansfield, M.A. Contreras, H.R. Moutinho, K. Ramananthan, R. Noufi, and M.M. Al-Jassim, Electrical conduction channel along the grain boundaries of $Cu(In,Ga)Se_2$ thin films, Appl. Phys. Lett. 102, 253905 (2013).
- D. Kuciauskas, A. Kanevce, J.M. Burst, J.N. Duenow, R. Dhere, D.S. Albin, D.H. Levi, and R.K. Ahrenkiel, Minority Carrier Lifetime Analysis in the Bulk of Thin-Film Absorbers Using Subbandgap (Two-Photon) Excitation. IEEE J. Photovolt. 3, 1319-1324 (2013).
- D. Kuciauskas, S. Farrell, P. Dippo, J. Moseley, H. Moutinho, J. V Li, A.M. Allende Motz, A. Kanevce, K. Zaunbrecher, T.A. Gessert, D.H. Levi, W.K. Metzger, E. Colegrove, and S. Sivananthan, Charge-carrier transport and recombination in heteroepitaxial CdTe. J. Appl. Phys. 2014, 116 123108 (2014).
- A. Kanevce, D. Kuciauskas, D.H. Levi, A.M. Allende Motz, and S.W. Johnston, Two dimensional numerical simulations of carrier dynamics during time-resolved photoluminescence decays in two-photon microscopy measurements in semiconductors, J. Appl. Phys., 118 045709 (2015).
- A. Allende Motz, J. Squier, D. Kuciauskas, S. Johnston, A. Kanevce, and D. Levi, Development of 2PE TRPL microscopy for lifetime and defect imaging in thin film PV materials and devices, IEEE PVSC42 Proceedings, in press (2015).

5. FY13-FY15 Special Recognitions, Awards, and Patents

- NREL ROI 13-047, Provisional Patent Application, March 2015, "Defect Characterization with thermoelectric cooling/heating", Jian Li, Dean Levi
- D. Kuciauskas, 2014 NREL President's award for "pioneering work applying nonlinear multiphoton techniques to revolutionize time-resolved photoluminescence measurements of carrier dynamics in PV materials."

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Colorado School of Mines	Golden, CO/ Jsquier@mines.edu	Support for graduate student and advisor/ Development of two-photon excitation microscopy.	158
Politecnico di Milano	Milano, Italy giorgio.ferrari@polimi.it	Design and fabrication of custom pre-amplifier for low-capacitance transient measurement for non-commercial SPM-based DLTS	56

Cell and Module Performance Development

Performing Organization: National Renewable Energy Laboratory

Key Technical Contact: Keith Emery (Primary Investigator), 303-880-2913, keith.emery@nrel.gov

Budget (FY13-FY15): $7.8M Agreement #: 25774

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1. Agreement Description and Motivation

The goal of this project is to position the SETP on a trajectory to meet the DOE SunShot Initiative goals by applying state-of-the-art current-versus-voltage and quantum efficiency measurements to support cell and module development that will drive improvements in the performance, reliability, cost, and manufacturability of photovoltaic (PV) materials, devices and systems. The politics of efficiency require that an independent laboratory perform these measurements with procedures that are accurate and traceable to recognizable standards for credibility. This task has provided a resource to the PV community for consultation on all aspects of I-V and QE measurements. NREL's expertise and capabilities span all technologies to facilitate research, development and testing, and evaluation in support of each of the PV technology research areas, including: CIGS, CdTe, CZTS, Silicon, Concentrator PV, Organic PV, Next-generation PV technologies, and Cell and Module Reliability R&D.

There is an increasing desire to reduce the uncertainty in the peak watt rating. Primary reference cells typically have uncertainties of 1% or less, which is the target uncertainty for secondary calibrations of cells and modules. This challenge is coupled with a demand for rapid turnaround. The increasing efficiency of multijunction concentrator cells adds additional uncertainty components related to concentrated light, linearity of the photo-current with irradiance, and spectral adjustability. This project analyzed all the significant sources of error related to PV power measurements and, as possible, reduced them by modifying existing hardware and procedures.

The final milestone for this agreement is to meet the other milestones and this has been accomplished.

2. Significant Achievements

- During the 3-year SunShot program the Provided DOE SunShot partners in the U.S. PV community over 21,000 measurements on 3600 samples. This allows meaningful independent comparisons of the efficiency between and within competing technologies. This allowed accredited U.S. based cell and module testing labs to have calibration traceability with the lowest possible uncertainty. The group also assisted numerous groups in reducing their measurement uncertainty by better understanding of sources of uncertainty, calibrations resulting in improved procedures.
- Eliminate the need for inaccurate temperature coefficients to correct the current for temperature. The procedure is mainly for calibration labs and requires the measurement of the spectrum and the QE as a function of temperature is known.
- Documented the errors in concentrating multijunction PV measurements when the spectrum cannot be adjusted and proposed a procedure to correct for this error when the spectrum can only be adjusted for 2 of the 3 or more junctions.
- Obtained ISO 9001 accreditation for all data reported to customers. The ISO 9001 quality system follows the ISO 17025 quality system, which has sample package limitations.
- Reduced the uncertainty in primary reference cell calibrations to below 0.9%. Reduced the uncertainty in module calibrations and verified by intercomparisons to be less than 2.5%. Reduced the cell uncertainty by using a more accurate reference cell.

3. Modifications and Remaining Challenges

- The scope was expanded to include obtaining and maintaining ISO 9001 accreditation for all data leaving the group. This supports the group's claim that all data is taken and released under the ISO 17025 quality system. The primary difference is a formal calibration certificate and uncertainty analysis is not required for work under the ISO 9001 banner.

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5. FY13-FY15 Special Recognitions, Awards, and Patents

- Keith Emery, William R Cherry Award, 2013.
- In 2014, Thomson Reuters listed Keith Emery as a highly cited researcher and one of the world's most influential scientific minds.
- In 2015, Thomson Reuters listed Keith Emery as a highly cited researcher.

6. University and Industry Partners

The Cell and Module Performance Development project did not collaborate with any external industrial or university partners.

Predicting Service Life for PV Modules

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: John Wohlgemuth (Primary Investigator), 303-384-7982, john.wohlgemuth@nrel.gov

Budget (FY13-FY15): $9.56 M Agreement #: 25812

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1. Agreement Description and Motivation

The objective of this project was to develop a set of accelerated stress tests that can provide a quantitative prediction of module service life in a variety of climates for different PV technologies and applications. This work was performed in conjunction with the International PV Quality Assurance Task Force (PVQAT). With this collaboration of many PV industry entities around the world, much more has been achieved than if the work were performed at only one or a few laboratories. This work was focused on developing a set of IEC standards to differentiate performance in multiple climates and applications.

Providing methodologies for quantitatively comparing the expected performance lifetimes between different commercially available module technologies will allow investors to better evaluate which module types to purchase for their systems. It will also reduce risk in the selection of modules, therefore reducing the cost to finance those systems. Multiple test sequences examining beyond the scope of IEC 61215 were being proposed in the industry at the start of this agreement, therefore we sought to minimize the cost of such evaluation using a single series of tests. By establishing a consensus set of tests, the amount of required accelerated testing may actually be reduced from the level it is at today, where each investor may require the module manufacturers to provide test data based on the investor's own set of preferred accelerated stress tests.

The final milestone for this agreement is to complete the research that convinces IEC National Committees to approve publication of IEC standards for climate specific lifetime testing of PV modules. This includes: identifying the needed UV dose for evaluating encapsulant discoloration and for verifying adhesion; determining the number of thermal cycles to eliminate solder bond and interconnect field failures; and demonstrating that leg 2 duplicates the types of delamination and corrosion that we see in the field. To be useful, these tests must induce failures in technologies that pass the current IEC 61215 but fail in the field in less than twenty-five years.

2. Significant Achievements

- Established a proposed Climate Specific Test Sequence. An IEC New Work item was approved and a Committee Draft was prepared and commented on.
- Technical Specification defining test methods for Potential Induced Degradation (PID) was published by IEC.
- Developed and published "Qualification Plus" to validate PV module reliability beyond the IEC 61215 qualification tests. At least one commercial product has been certified to "Qualification Plus".
- Technical Specification for the Guidelines for PV module manufacturing Quality Management System is under development. It has been submitted to IEC for the final vote before publication.
- The 2nd edition of the IEC module safety test was unanimously approved by the National Committees. A number of clarification questions will be answered before it can be published.
- The "IECRE Conformity Assessment System" was formally approved by IEC, the governing body for the system was established, and it was tasked with preparing the rules for operating the IECRE system.
- Established an international effort to develop a unified approach to testing for module performance in hot climates.
- A set of standards for module materials (encapsulants, backsheets and edge seals) is progressing through the IEC system toward publication.
- Developed a thermal/mechanical model for PV modules and used it to predict the equivalent stress levels between field exposure and thermal cycling. The worst case evaluated so far indicated than 25 years in Chennai, India

can be simulated by 625 thermal cycles. This modeling also indicates that using a higher upper limit for the thermal cycling results in significantly greater stress. A New Work item on thermal cycling has been prepared for submission to IEC as part of the Climate Specific Test Sequence.

- Conducted an inter-laboratory experiment evaluating the loss of optical transmittance in encapsulants as a function of UV, temperature and humidity. The results have been published and a New Work item on UV testing of PV module materials has been prepared for submission to IEC as part of the Climate Specific Test Sequence.
- Conducted an inter-laboratory experiment to evaluate the impact of UV exposure, temperature and humidity on the adhesion between the encapsulant and front glass. The results are temperature dependent. At higher exposure temperatures (90°C) the adhesion falls with UV exposure, but then stabilizes at ~ 40 % of the initial value, which appears adequate for long term module survival. The results will be used to redefine the proposed Leg 2 test to cause delamination.

3. Modifications and Remaining Challenges

Discovery experiments on Leg 2 of the Climate Specific Test Sequence have not been successful at causing delamination in modules that successfully pass the qualification tests. Since we have not been able to demonstrate Leg 2, the international community indicated that we were not ready to submit a CDV (Committee Draft for Vote) for the Climate Specific Test Sequence.

4. FY13-FY15 Publications

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- Michael D. Kempe, Dylan L. Nobles, Mark D. Weigel, Alan K. Nachtigal, Mark A. Roehrig, Tracie J. Berniard, Joseph C. Spagnola, an Charlene M. Schubert, "Evaluation of the Durability of Flexible Barrier Materials", 42nd IEEE PVSC, New Orleans, 2015.
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5. FY13-FY15 Special Recognitions, Awards, and Patents

None

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
University of Central Florida	Coco, Fl. Neelkanth Dhere dhere@fsec.ucf.edu	Deploy and periodically inspect and measure modules made with different encapsulants.	24
University of Central Florida	Coco, Fl. Stephen Barkaszi barkaszi@fsec.ucf.edu	Deploy and periodically inspect and measure modules on a high voltage test bed.	16
ASU	Mesa, AZ Mani TamizhMani manit@asu.edu	Analysis of Arco modules – field deployed for 27 years versus stored in shed.	30
MIT	Cambridge, MA Tonio Buonassisi buonassi@mit.edu	Analysis of Arco modules – field deployed for 27 years versus stored in shed.	30
Stanford University	Stanford, CA Reinhold Dauskardt rhd@stanford.edu	Analysis of Arco modules – field deployed for 27 years versus stored in shed.	30

Quantifying Risk through Bankability Reports

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Sarah Kurtz (Primary Investigator), 303-384-6475, sarah.kurtz@nrel.gov

Budget (FY13-FY15): $3M Agreement #: 25810

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1. Agreement Description and Motivation

This project seeks to improve bankability of PV by documenting PV performance on a broad scale and by developing tools to characterize energy produced and module degradation rates. The work spans from collecting high-accuracy data to doing statistical evaluations of data others have reported. Special emphasis was placed on understanding degradation rates: both what is being reported in the literature and how to reduce the uncertainty in measuring degradation, especially in a short time. Emphasis was also placed on writing standards that are of high priority to PV customers.

2. Significant Achievements

- Updated the review paper on "Photovoltaic degradation rates" - the original paper has been highly cited by the community.
- Instrumental in creating international specifications for measuring the power and energy generated by PV plants in order to facilitate writing of performance guar.
- Published set of papers summarizing the data from 1.7 GW of installations demonstrating that the majority of systems installed under this program function as expected.
- Published study showing the lack of degradation of a CPV module deployed for ~ 6 years.
- Defined the length of data collection needed to calculate a degradation rate with a given uncertainty as a function of the quality of the data.
- Completed instrumentation of GSA system in North Carolina.
- Lay technical groundwork for general approach to a standard method for determining degradation rates for PV systems.
- Published IEC 62817 (Design qualification for Solar Trackers) enabling higher reliability for both flat-plate and CPV trackers.

3. Modifications and Remaining Challenges

- The installation of the hardware for monitoring the GSA project in North Carolina was delayed by needed arrangements with GSA.

4. FY13-FY15 Publications

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- Grenko, Brian, Adie Kimber, and Sarah R. Kurtz. "Rating PV Modules for Field Performance," *Solar Pro*, October/November 2014.
- Hashimoto, J. Y. Xue, K. Otani, M. Muller, and S. Kurtz "Performance of Grid-connected Conventional Flat-plate and Concentrator Photovoltaic Systems in Japan" ICEE 2014.
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5. FY13-FY15 Special Recognitions, Awards, and Patents

None

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Ryan Smith		Support in software development and data handling	150

Emerging Technology Characterization

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Bill Marion (Primary Investigator), 303-384-6793, bill.marion@nrel.gov

Budget (FY13-FY15): $3.0M Agreement #: 25809

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1. Agreement Description and Motivation

This work sought to remove barriers due to uncertainties associated with the performance of emerging technologies. The emerging technologies addressed were thin-film and concentrator PV modules. Key elements of the work were: (a) collecting outdoor performance data, (b) developing methods and procedures to accurately determine the performance of thin-film and concentrator PV modules at standard test conditions (STC), and (c) developing methods and procedures to translate performance from STC to outdoor conditions in order to estimate the energy production of a PV system.

The final milestones for this agreement were:

- Successful completion of Task 3, resulting in the understanding of best cell structure designs for two locations; the CPV system characteristic data required for predicting CPV system level performance identified/verified within 5%, and quantifying the impact on ratings due to procedural differences in testing.
- Successful completion of Task 4, resulting in error associated with temperature during light soak is determined; alternate method demonstrated for successful and unsuccessful procedures; and accuracy of stabilization methods are compared to each other and for predicting outdoor performance.

2. Significant Achievements

- Completed and made publicly available a first-of-kind PV module performance and meteorological data set encompassing three climates (Cocoa, FL; Eugene, OR; and Golden, CO) and all flat-plate PV technologies.
- Analyzed the PV module data and showed significant variation in energy production, due to both the site-to-site differences in reference yield and the PV module characteristics, with the best performing PV modules at Cocoa, FL having final PV yield values nearly 60% greater than the lowest performing PV module at Eugene, OR.
- Optimized a procedure for translating field measurements to CPV rating conditions and reduced the uncertainty from 10% to the goal of 5%, solving a critical technical problem for rating CPV modules. The optimized procedure was submitted into the draft IEC 62670-3.
- Identified the essential characteristic data for predicting CPV module performance and for quantifying the impact of cell structure on CPV module energy production.
- Used essential characteristic data for CPV module to model the performance of a CPV system and resultant error was less than 5%.
- Developed a stabilization method for the indoor measurement of CIGS PV modules and validated with round robin testing. Achieved the measurement goal of reducing the variability between labs from 8% to 3%.
- Determined that the measurement at STC of CdTe and CIGS PV module is sensitive to the temperature of the PV modules during the light soak preconditioning step.
- Improved on the draft IEC 61215 to minimize meta-stability effects for reducing the uncertainty of measurements at STC for CdTe and CIGS PV modules.

3. Modifications and Remaining Challenges

Beginning in FY2014, some of the original planned work was moved to another agreement and other work was not funded. Work related to PV module level power electronics and baseline testing of PV modules deployed at SolarTac was moved to the Regional Test Center agreement. Work related to the validation of public and commercial PV module power or energy models/algorithms/procedures using the comprehensive PV module I-V curve data set was not funded. Work related to the development and validation of a model/method to correct the performance of any flat-plate PV technology for variations in the spectral distribution of solar irradiance from variations in air mass, aerosols, precipitable water vapor, and clouds was not funded. Funding amounts were $1,707K in FY2013, $823K in FY2014, and $480K in FY2015.

4. FY13-FY15 Publications

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- T.J. Silverman, G Deceglie, B Marion, S Cowley, B Kayes, S Kurtz, “Outdoor Performance of a Thin-Film Gallium-Arsenide Photovoltaic Module”, Proceedings of the 39th IEEE PVSC, Tampa, 2013.
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- M. G. Deceglie, T. J. Silverman, B. Marion, S. Kurtz, “Temperature-Dependent Light-Stabilized States in Thin-Film PV Modules,” in Proceedings of the 41th IEEE PVSC, New Orleans, 2015.
- M. G. Deceglie, T. J. Silverman, B. Marion, S. Kurtz, “Robust measurement of thin-film photovoltaic modules exhibiting light-induced transients,” SPIE Conference, San Diego, CA, 2015. http://www.nrel.gov/docs/fy15osti/64769.pdf
- M. G. Deceglie, T. J. Silverman, B. Marion, S. Kurtz, “Real-time Series Resistance Monitoring in PV Systems without the Need for IV Curves,” accepted for publication in the IEEE Journal of Photovoltaics on August 20, 2015.
- M. Muller, B. Marion, S. Kurtz, K. Ghosal, S. Burroughs, C. Libby, ”A Side-by-Side Comparison of CPV Module and System Performance”, submitted September, 2015 for publication in Progress in Photovoltaics.

5. FY13-FY15 Special Recognitions, Awards, and Patents

Patent Application

- Deceglie, M. G., “Real-time Series Resistance Monitoring in Photovoltaic Systems,” April 10, 2015.

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
University of Oregon/Frank Vignola	fev@uoregon.edu	Measurements for Validating the Accuracy of Models for Predicting the Performance of PV Modules	108
CFV Solar Test Laboratory/Steve Dudden	steve.dudden@cfvsolar.com	PV Module Characterization and IEC 61853 Measurements	57

Regional Test Center Support – NREL Site

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Chris Deline (Primary Investigator), 303-384-6359, chris.deline@nrel.gov

Budget (FY13-FY15): $2.3M Agreement #: 29185

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1. Agreement Description and Motivation

The DOE seeks opportunities to increase US manufacturing and deployment of PV. A key barrier to the success of new PV technologies is demonstrating the performance and reliability at a level that gives investors adequate confidence for large-scale investments. The DOE created the Regional Test Centers (RTCs) to help companies bridge this "Valley of Death" by providing infrastructure to rapidly test new products. In particular, the NREL portion of the RTC Support agreement covers the management of the NREL RTC site, including acquisition of new partners, deployment of 2 new systems per year, and monitoring existing systems for data quality and system health. The final milestone for this agreement is to provide the proposed support for each RTC partner, positioning each partner to achieve bankability. Specifically, 100% of reports promised to RTC partners in their test plans must be completed in a timely way. Data quality reviews must show that 95% of data were available.

2.1 Significant Achievements (FY15)

- R&D 100 award finalist: "Maxim Integrated Solar Cell Optimizer" (NREL and Maxim joint application). *Impact: Demonstration and promotion of innovative US partner technology*
- Shared Resources CRADA partnerships signed in FY15: SolarWorld, Silevo, Prism Solar, HST Solar. Total CRADA value: $1.45M. *Impact: SunShot goals of supporting US manufacturing and leveraging private capital– these field deployments help improve the bankability and reduce uncertainty of partner technologies.*
- Invited talk at Trina Solar for their annual R&D meeting, topic of embedded PV power electronics. *Impact: Promoting US RTC manufacturing partners to international customers.*
- Degradation rate analysis for TenKSolar product technology. Used by NRG energy to obtain project financing, based on -0.5%/year degradation rate assessment. *Impact: Improving the bankability and reducing uncertainty of novel US manufacturing and technologies.*
- Completed the Stion RTC materials analysis project by assessing the long-term stability of various module components under UV exposure. *Impact: Technical assessment of component reliability and direct company support.*
- Program management metrics: 99% average data availability (success); 100% delivery of reports and other deliverables within 1 month of due date (21 out of 21); CRADA's negotiated within 4 months of partner acceptance to the RTC program (2 out of 3).
- Snow loss and soiling rate systems installed, enabling performance losses from these factors to be monitored within 2% (success).
- Company surveys indicate that 100% of respondents are satisfied with the RTC support they have received (success).
- Development of a new PV deployment site on NREL's main campus for local installation of new RTC partner systems, up to 160 kW.

2.2 Significant Achievements (FY13-FY14)

- Deployment of first two (45 kW) RTC installations (Maxim and Heliovolt). Development of RTC technical validation procedures and management processes.

3. Challenges and Modifications

The proposed work was conducted to plan, although progress can be slower than anticipated at times. Some project challenges include:

- CRADA agreements are frequently delayed because of legal review and negotiations.
- Partner test plans can require many iterations if testing requirements change, or if the RTC

partner is unclear on the technical merits of the validation work.

- Working with small startup companies carries a risk, if the company doesn't stay solvent long enough to complete a demonstration project.

Despite these risks and challenges, the ability to complete the program management metrics, and continue to bring new RTC partners to the program was successfully demonstrated in FY15.

4. FY13-FY15 Publications

- C. Deline, B. Sekulic, J. Stein, S. Barkaszi, J. Yang, S. Kahn, "Evaluation of Maxim Module-Integrated Electronics at the DOE Regional Test Centers", 40th IEEE PVSC 2014 http://www.osti.gov/scitech/biblio/1136207
- C. Deline, "Opportunities and challenges in the development of smart PV systems", presented at 2015 Trina State Key Laboratory annual meeting, Changzhou, China June 25-26, 2015. NREL- PR-5J00-64525.
- RTC reporting documents (unpublished):
    - Heliovolt Performance and degradation rate assessment (5/2014)
    - Soitec Module outdoor performance characterization (with Sandia, 4/2014)
    - Maxim NREL capacity test report (8/2014)
    - Maxim NREL quarterly performance report (2/2015)
    - Stion Materials selection following accelerated UV stress (5/2015)
    - TenKSolar Performance assessment and degradation rate analysis (7/2015)
    - Maxim FSEC capacity test report (8/2015)
    - TenKSolar Analysis of module construction and failure modes (8/2015)

5. FY13-FY15 Special Recognitions, Awards, and Patents

- R&D 100 award finalist: "Maxim Integrated Solar Cell Optimizer" (NREL and Maxim joint application)

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY13-FY15. Note: CRADA amounts reflect the total project value including DOE's share and the partner's share.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	Agreement value ($K)
Heliovolt	No longer in business	CRADA- Validation of Heliovolt modules at the RTCs	1115
Maxim	Seth Kahn Seth.Kahn@maximintegrated.com	CRADA – Validation of Volterra modules at the RTCs	1400
HST Solar	Rudy Roy rudy@hstsolar.com	CRADA- HST Solar's Advanced Mounting Systems at the RTCs	860
Prism Solar	Paul Hauser p.hauser@prismsolar.com	Bailment- Prism module loan for RTC assessment	1
SolarWorld	Olson, Eric Eric.Olson@SolarWorld.com	CRADA- Validation of SolarWorld Modules at the NREL RTC site	328
Silevo	Christoph Erben cerben@solarcity.com	CRADA- Validation of Silevo Modules at the NREL RTC site	264

NREL Regional Test Center Research

Performing Organizations: National Renewable Energy Laboratory

Key Technical Contacts: Chris Deline (Primary Investigator), 303-384-6359, chris.deline@nrel.gov

Budget (FY14-FY15): $1.5M Agreement #: 24371

1. Agreement Description and Motivation

A key barrier to the success of new PV technologies is demonstrating the performance and reliability at a level that gives investors adequate confidence for large-scale investments. The DOE created the Regional Test Centers (RTCs) to help companies bridge this "Valley of Death" by providing infrastructure to rapidly test new products. Two research projects are described here that depend on the RTCs to generate data and methodologies, but will have an impact relevant to the wider industry:

1. Development of methods for assessing the performance and economic value of module-embedded power electronics. This work will have immediate application to the Maxim RTC installations, and is designed to update associated standards defining performance benefits of distributed electronics.
2. Continuing research on an issue that was identified with a current RTC partner. This issue is a failure mechanism that is not detected by IEC 61646, the thin-film module qualification test. Other thin-film companies have reported failures that may be related and are also missed by IEC 61646. We propose to define a technical basis for creating a standalone reliability screening test so that these types of failures are identified in a more timely way.

The final milestones for this agreement are:

1. To provide updated partial shade performance models to the SAM team, enabling distributed electronics to be evaluated within 2%;
2. To develop an accelerated method for detecting thin-film failure from partial shade.

2.1 Significant Achievements

- Development and transfer of partial shading PV performance models to the community:
    - 2 Journal and 3 Conference publications.
    - Open source software made available.
    - Model integrated into NREL's SAM performance assessment tool.
    - Model shared directly with commercial partners (SunRun & SunPower) for use in their proprietary PV model tools.

  Impact: enable the performance advantages of distributed power electronics (microinverters and power optimizers) to be correctly assessed under typical residential conditions.
- Standard shade test methodology for side-by-side comparison of string inverter vs. sub-module embedded power electronics (Maxim)

  Impact: Provide the methodology to evaluate emerging embedded sub-module power electronics under closely-spaced deployment scenarios like commercial rooftop.
- Reliability screening test developed for monolithic thin-film modules.
    - Premature field failure was detected due to deposition / illumination nonuniformity.
    - Formal method developed to identify the onset of this particular reliability issue and ways to identify modules resistant to partial shade damage.
    - Results and method transmitted to the community via 2 journal and 2 conference papers.

  Impact: Enable monolithic CIGS manufacturers to assess permanent performance losses under real-life shading conditions, and begin engineering more durable designs.
- Development of low-cost outdoor imaging methods for field damage assessment.

  Impact: A $500 imaging tool was found to take outdoor daytime PL images equal to a $50,000 InGaAs camera. This low cost point could result in much more widespread use of optical damage imaging for fielded cSi and thin-film systems. Although long exposure times currently limit the usefulness of the method.

2.2 Program milestones

- Provide the community with a standard shading performance test method to accurately assess the ability of module-embedded DC-DC converters to increase the energy density of PV installations (complete, journal article submitted).
- SAM shading models are updated to enable the annual energy benefits of distributed electronics to be evaluated within 2% (complete).
- Develop accelerated method for detecting thin-film failure that has been observed in the field and draft a procedure to be proposed as part of a standard qualification test, or as a standalone reliability assessment procedure (complete).

3. Challenges and Modifications

- An early go/no-go milestone was set in Feb. 2014 to assess the value of the thin-film reliability work to industry, and to revise the initially proposed experiment and research plan. Following industry input, and with expert review by our DOE technical managers, we were able to craft an appropriate work plan that came to a successful conclusion.

4. FY14-FY15 Publications

- Alex Hanson, Chris Deline, Sara MacAlpine, Jason Stauth, Charles Sullivan, "Partial-Shading Assessment of Photovoltaic Installations via Module-Level Monitoring", IEEE Journal of PV **4** pp. 1618-1624, 2014. http://www.nrel.gov/docs/fy15osti/63765.pdf
- Carlos Olalla, Chris Deline, Dan Clement, Yoash Levron, Miguel Rodriguez, Dragan Maksimovic, "Performance of Power-Limited Differential Power Processing Architectures in Mismatched PV Systems." IEEE Transactions on Power Electronics **30** pp. 618-631, 2015
- Tim Silverman, Mike Deceglie, Xingshu Sun, Rebekah Garris, Muhammad Alam, Chris Deline, Sarah Kurtz, "Thermal and electrical effects of partial illumination in monolithic thin-film photovoltaic modules", IEEE Journal of Photovoltaics JPV-2015-06-0259-PVSC.R1 (in press), 2015. http://www.nrel.gov/docs/fy15osti/64448.pdf
- Chris Deline, "Inverters, power optimizers, micro-inverters" (ch. 11.2). in Photovoltaic Solar Energy: From Fundamentals to Applications, Wiley and Sons, in press
- S. MacAlpine, C. Deline, "What are some recommendations for modeling microinverters in PVWatts?" chapter in Renewable Energy Handbook, 2016 edition, Solar Power World, 2015. http://www.solarpowerworldonline.com/2015/09/what-are-some-recommendations-for-modeling-microinverters-in-pvwatts/
- C. Deline, B. Sekulic, J. Stein, S. Barkaszi, J. Yang, S. Kahn, "Evaluation of Maxim Module-Integrated Electronics at the DOE Regional Test Centers", 40th IEEE PVSC 2014. http://www.osti.gov/scitech/biblio/1136207
- Sara MacAlpine, Chris Deline, "Simplified Method for Modeling the Impact of Arbitrary Partial Shading Conditions on PV Array Performance", 42nd IEEE PVSC, 2015. http://www.nrel.gov/docs/fy15osti/64570.pdf
- Xingshu Sun, John Raguse, Chris Deline, Tim Silverman, Muhammad Alam, "A physics-based compact model for CIGS and CdTe solar cells: from voltage-dependent carrier collection to light-enhanced reverse breakdown", 42nd IEEE PVSC, 2015.
- Kate Doubleday, Chris Deline, Carlos Olalla, Dragan Maksimovic, "Performance of Differential Power-Processing Submodule DC-DC Converters in Recovering Inter-Row Shading Losses", 42nd IEEE PVSC, 2015. JPV journal article submitted.
- Tim Silverman, Michael Deceglie, Chris Deline and Sarah Kurtz, "Partial shade stress test for thin-film photovoltaic modules", 2015 SPIE Optics + Photonics, San Diego, CA, 2015. http://www.nrel.gov/docs/fy15osti/64456.pdf
- Chris Deline, "Opportunities and challenges in the development of smart PV systems", Trina State Key Laboratory annual meeting, Changzhou, China, June 2015.
- Chris Deline, "Performance Modeling and Testing of Distributed Electronics in PV Systems", IEEE Applied Power Electronics Conference, Charlotte, NC March 2015. http://www.nrel.gov/docs/fy15osti/64167.pdf
- K Ramanathan, L Mansfield, R Garris, C Deline, T Silverman, "Processing and Device Oriented Approach to CIGS Module Reliability", NREL PV Module Reliability Workshop, Golden, CO 2015. http://www.nrel.gov/docs/fy15osti/63826.pdf
- Steve Johnston and Tim Silverman, "Photoluminescence and Electroluminescence Outdoor Module Imaging", NREL PV Module

Reliability Workshop, Golden, CO 2015. http://www.nrel.gov/docs/fy15osti/64438.pdf

- C. Deline, J. Meydbray, and M. Donovan, "Photovoltaic Shading Testbed for Module-Level Power Electronics: 2014 Update", NREL/TP-5J00-62471, 2014. http://www.nrel.gov/docs/fy14osti/62471.pdf
- Sara MacAlpine, Chris Deline, "Modeling Microinverters and DC Power Optimizers in PVWatts", NREL/TP-5J00-63463, 2015. http://www.nrel.gov/docs/fy15osti/63463.pdf
- Chris Deline, "Renewable Energy, Photovoltaic Systems Near Airfields: Electromagnetic Interference", NREL/TP-5J00-63310, 2015. http://www.nrel.gov/docs/fy15osti/63310.pdf

5. FY13-FY15 Special Recognitions, Awards, and Patents

- R&D 100 award finalist: "Maxim Integrated Solar Cell Optimizer" (NREL and Maxim joint application)
- Provisional Patent no. 62/218,104 "PV Module-level remote safety disconnect" by C. Deline filed 9/14/2015

6. University and Industry Partners

The following organizations partnered in the project's research activities during FY14-FY15.

Organization/ Principal Investigator	Location/E-mail	Description/Title of Research Activity	($K)
Purdue University	alam@purdue.edu	Development of CIGS compact device model to aid in 2D partial shade response simulations	78