

Solid-State Lighting Program: LED Materials and Devices R&D Meeting

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Table of Contents

1	Introduction.....	7
2	Key Findings.....	7
2.1	Application Considerations	8
2.2	Materials and Device Science.....	8
2.3	Device Concepts	8
2.4	Optical Down-Converters.....	8
	Appendix A: Participant Presentations.....	10
	Hee Jin Kim, Lumileds: Efficient Green and Yellow LEDs for Solid-State Lighting Applications....	10
	Yuya Harada, Nichia: Technologies for Social Contributions	10
	Jim Speck, University of California (UC), Santa Barbara: Differential carrier lifetime measurements	10
	Siddharth Rajan, Ohio State University: Efficient Green and Yellow LEDs for Solid-State Lighting Applications.....	11
	Zetian Mi, University of Michigan: A Nanoscale Approach to Overcome Challenges of LED Lighting	11
	Jim Murphy, GE Research: Phosphors for Next Generation Solid-State Lighting/Displays	11
	Jon Schuller, University of California, Santa Barbara: GaN Light Emitting Quantum Well Metasurfaces.....	11
	John Epler, Lumileds: Direct Red Development: Improved AlInGaP VTF and Novel Flip Chip Architecture	12
	Kirstin Alberi, National Renewable Energy Laboratory: Red and Amber LEDs, Leveraging Multijunction PV Performance Improvements.....	12
	Ilan Jen-La Plante, Nanosys: Quantum Dot Downconverters for Next-Generation Solid-State Lighting, Challenges and Opportunities	12
	Emory Chan, Molecular Foundry: Automated Discovery of Nanophosphors for Solid State Lighting Guided by Machine Learning	12

1 Introduction

On August 23, 2022, 12 experts from industry, national laboratories, and universities gathered at the invitation of the Department of Energy (DOE) Solid-State Lighting (SSL) Program to identify critical research and development (R&D) topic areas in light emitting diode (LED) based lighting.

The meeting was held virtually. The meeting commenced with "soapbox" presentations where each participant gave a short presentation describing what they believed to be the key technology challenges in SSL over the next three to five years. This was followed by a general discussion of the most critical technology challenges facing the industry today.

The meeting format provided an opportunity for the LED experts to exchange ideas and explore collaborative research concepts.

This report summarizes the outcome of the discussions on critical technology challenges and identifies corresponding R&D tasks. Outlines of the participants' soapbox presentations and related remarks are included in Appendix A of the report.

2 Key Findings

All of the researchers suggested ongoing R&D in LED materials and device topics. While LED technology has rapidly improved, there are still opportunities to improve efficiency and reduce performance trade-offs. At peak performance conditions and typical LED device architectures, blue LED efficiency has plateaued at exceptionally high levels. However, breakthroughs in green and amber LED performance, droop mitigation, optical control, and optical down-converter materials could enable the next level of efficiency, energy savings, and functionality. These advancements would broaden the range of conditions and architectures that could exhibit the highest levels of power conversion efficiency, luminous efficacy, and energy savings.

The attendees noted that current droop is still a major limitation for LED performance, limiting cost, efficiency, and light output levels. Current droop also limits the practical luminance, optical control, and the range of practical lighting device architectures. Green LED performance still greatly lags blue LED performance, requiring optical down-conversion to achieve white light emission. This limits the technology's color control, color options, and potential efficiency. Improvements to optical down-converters can enable improvements in luminous efficacy, new color options, and new device architectures. Conventional LEDs are also limited in optical control and luminance not just in terms of light output per area but also in the optical distribution achievable at the LED chip and package level.

The topics put forth by the attendees reflect the rapid advancement in fundamental scientific understanding enabled by new LED technology and describe ongoing improvements to this understanding. Already, advancements in LED technology and materials have led to advancements in understanding of semiconductor optoelectronic materials, carrier dynamics, optical down-conversion physics, and nano-scale physical phenomena associated with these devices. It is highly likely that continued R&D on LED devices and materials will result in additional scientific breakthroughs within the topic of semiconductor optoelectronics.

The meeting format encouraged attendees to participate and present their perspectives on critical R&D challenges. In the discussions that followed the soapbox presentations, several recurring themes arose regarding research areas that could lead to significant breakthroughs in LED performance (in more detail in the sections below):

- Application Considerations
- Materials and Device Science

- Device Concepts
- Optical Down-Converters

2.1 Application Considerations

LED device and material development is driven by new understanding in applications such as displays, human physiological responses to light (health and wellness), horticultural lighting, germicidal ultraviolet light for disinfection, and improved color quality for general lighting applications. These new applications benefit from new device architectures and metrics to characterize the intended function of the light beyond traditional photopic lighting metrics with their well-understood limitations. Ongoing research in these and other new applications will continue to drive the development of improved LED materials and devices. This development will address the new application understanding and corresponding new performance requirements. The wide range of colors, intensity, optical control, and form factors enabled by LED technology enables these new features and application benefits. There will be a virtuous cycle of newly enabled applications and benefits with LED lighting, development of improved LED devices, and research to better understand the newly enabled applications and benefits which again enables new applications and benefits. Since LED lighting is still somewhat new, most of these new applications and benefits are not yet well addressed.

2.2 Materials and Device Science

LED technology has matured rapidly. However, advancements in scientific understanding of LED devices and materials are ongoing, enabled by new and better devices, materials, and characterization techniques. The relationship between V-pit defects and forward voltage is being explored such that V-pit defects can be engineered to improve device performance. Improved device performance models are being developed and validated, which provide insight into the underlying physical mechanisms within LEDs (and other types of opto-electronic devices). Performance differences in LEDs with different emission wavelengths are being explored to better understand the underlying phenomena that affect performance. These include polarization fields within the LED crystal material, defects, and radiative and non-radiative recombination rates. Improved understanding of the physical mechanisms of LEDs can enable performance improvements (particularly for green emitting devices), high current operation, and high luminance operation.

2.3 Device Concepts

Improved understanding of the underlying physical mechanisms and efforts to address new lighting applications drive the development of new LED device concepts. Researchers are pursuing LED devices with tunnel junctions. These would reduce droop (high current density efficiency reduction) and result in devices with higher luminance levels. These would also increase light output per device area, which would further reduce the cost of LEDs. The use of metasurface optics with LEDs could increase radiance (or luminance) by enabling more directional light output from the LED surfaces. Metasurfaces are phased array passive nanoscale optical elements that could be deployed on the surface of LED chips. Preliminary data on the use of metasurfaces on LEDs show little to no reduction in external quantum efficiency. Research in nanostructure LEDs is also still being pursued. Nanostructured LEDs may be able to remove some of the limitations of planar LEDs such as crystal defects and polarization fields. All of these device concepts demonstrate that continued advancements in basic LED device structures are possible and enabled by ongoing advancements in scientific understanding. This could result in improved LED efficiency, luminance, optical control, or other features.

2.4 Optical Down-Converters

Phosphors are optical down-converters that are found in all white LEDs. Phosphor down-conversion properties have a heavy influence on white LED luminous efficacy, color quality, lifetime, and maximum operating conditions. In particular, red phosphor materials in white LEDs can require a strong trade-off between luminous efficacy and color fidelity as expressed by CRI or IES TM-30 R_f. Certain red phosphor materials can

minimize this tradeoff with optimized emission wavelength and spectral width. However, these narrow red emitting phosphors can be limited by other performance factors such as absorption and various quenching mechanisms. Researchers are working to reduce these detrimental factors to get the full benefit of improved luminous efficacy and color fidelity.

Quantum dots are another option for use as optical down-converter materials. They may be able to provide engineered emission color options, which would enable more precise spectral engineering of the LED emission. Quantum dots suffer from various quenching mechanisms, environmental stability concerns, and concerns over the inclusion of heavy metals. Cadmium-containing quantum dots have demonstrated the best efficiency and longest lifetime. Research is underway to improve cadmium-free quantum dots and understand the underlying physical mechanisms of the cadmium-containing materials. Since quantum dots are synthesized with chemical processes, innumerable chemical combinations could be used in their synthesis. Researchers are developing machine learning-driven combinatorial automated synthesis techniques to explore the materials and synthesis parameter space and to understand relationships between synthesis, materials, and ultimate quantum dot performance as an optical down converter.

Appendix A: Participant Presentations

Hee Jin Kim, Lumileds: Efficient Green and Yellow LEDs for Solid-State Lighting Applications

Hee Jin Kim, Senior Director for Epitaxy Technology R&D at Lumileds, discussed various aspects of the DOE funded project to improve the efficiency of green and amber LEDs. She compared the status and performance of state-of-the-art green LEDs to DOE SSL targets. She noted that commercialized green LEDs typically have either low forward voltage (V_f) or good external quantum efficiency (EQE) droop, but not in the same device. There are various engineering methods to increase overall efficiency and mitigate the trade-off between EQE and V_f at higher current density operation. V-pit engineering was evaluated to reduce forward voltage, but at higher current density the overall performance benefits and trade-offs were found to be unfavorable. Currently, cascade LEDs are being pursued as part of the project to mitigate current droop. An industry-academia collaboration is working on this project, including:

1. University of Michigan's model that predicts the changes in spectral properties with carrier density based on principles,
2. Work at University of New Mexico and Sandia National Laboratory that established that longer radiative lifetime explains the internal quantum efficiency (IQE) deficit at longer wavelength, and
3. Tunnel junction and novel LED designs by Ohio State University.

Progress has also been made on indium-rich InGaN quantum wells (QW). This was done with the same simulation technology developed at the University of Michigan. It was found that narrow well width can provide higher recombination and improved droop properties. However, this requires very high indium concentrations.

Yuya Harada, Nichia: Technologies for Social Contributions

Yuya Harada, Technical Support Manager at Nichia America Corporation, considered the impact of LED lighting and R&D directions that the industry should consider moving forward. The most apparent reasons for switching from conventional lighting sources to LEDs are cost and efficiency. But in recent years, there has been more discussion of the new values LED technology can provide, including health and light quality. The social contributions of lighting can be split into three categories: wellness, delight, and sustainability. These can be supported with continuing LED development. For example, wellness could be achieved through human centric lighting and disinfection, while delight can be provided through high quality of light. Sustainability requires reduction in energy consumption (source luminous efficacy, improved light delivery, sensors, and controls) and environmentally friendly designs (reducing fixture size, material consumption, etc.). Two specific ideas were presented for consideration. First was super high luminance LEDs for "invisible lighting", allowing for compact designs with fewer resources and less installation costs. Alternatively, super low luminance LEDs can be used for "non-bright lighting" to provide specialized, distributed, and glare-free lighting for more human centric illumination. The social contributions of lighting cannot be realized by LED manufacturers alone, but by increasing awareness and education overall.

Jim Speck, University of California (UC), Santa Barbara: Differential carrier lifetime measurements

Jim Speck, Professor at UC Santa Barbara, discussed screened versus unscreened quantum wells to see the role of polarization on overall LED device efficiency. Small-signal modulation of simple LED device structures enables evaluation of the frequency response of the LEDs. This allows for decoupling of carrier lifetime and carrier transport effects. Studies were performed on single quantum well LEDs emitting at violet, blue, and green wavelengths which reflect different indium levels in the quantum wells. These devices were further compared with and without doped barriers to see the effect on internal electric fields. Doping the barriers improved radiative recombination efficiency in all of the devices, but induced a small increase in non-radiative defect recombination. The results showed the isolated levels of defect recombination, radiative recombination,

and Auger recombination so that the impacts of device structure design and synthesis can be more clearly evaluated.

Siddharth Rajan, Ohio State University: Efficient Green and Yellow LEDs for Solid-State Lighting Applications

Siddharth Rajan, Professor at Ohio State University, discussed a joint project with Lumileds that aims to find new approaches to improve long wavelength LED efficiency. Tunnel junctions (TJ) provide flexibility in designing LEDs by cascading multiple active regions to circumvent efficiency droop problems. While tunnel junction devices show scaled optical output (2X output for 2 tunnel junction devices), voltage is much more than doubled, creating an unfavorable efficiency trade-off. Work is necessary to improve tunnel junction voltage performance, particularly related to p-type GaN material activation. Increasing p-type activation temperature reduces voltage but degrades the device EQE. An intermediate temperature allowed the team to reduce voltage drop. MOCVD TJ-LEDs are approaching voltage drop of optimized non-TJ LEDs with improvement in sidewall annealing conditions. Improved p-type activation techniques enable the possibility of p-type down device structure using a tunnel junction. While the p-type down architecture is showing promise, the optical performance of multi-quantum well devices for p-down structures is not as good as p-up, and will require additional research and understanding.

Zetian Mi, University of Michigan: A Nanoscale Approach to Overcome Challenges of LED Lighting

Zetian Mi, Professor at the University of Michigan, explained that the challenge for red and green InGaN devices is that when shrinking the device size, efficiency drops considerably. A nanocrystal-based structure with a bottom-up approach to grow the nanostructure can be used. This approach allows higher incorporation of indium in the active regions to help achieve high efficiency green, amber, or red emission from an InGaN QW or quantum dot (QD) emitter. A major challenge for nanoscale approaches is surface recombination. This was addressed by using an aluminum rich AlGaIn shell. This can suppress non-radiative surface recombination, improving device performance. A unique benefit offered by nanocrystals is that one can achieve RGB in the same wafer in a single growth step. This bottom up approach can also be patterned into optical arrays, which narrows down the emission spectrum to achieve a spectral linewidth of 4 nm. The nanowire approach for submicron LED device on wafer measurement gives 10% EQE. This is the most efficient submicron green LED currently demonstrated.

Jim Murphy, GE Research: Phosphors for Next Generation Solid-State Lighting/Displays

Jim Murphy, Senior Scientist at GE Research, argued that the phosphor on-chip design will continue to dominate the SSL industry. The display industry continues to grow, and the trend is moving towards smaller sized LEDs. Mini-LEDs are already on the market, and micro-LEDs are soon to be adopted. This trend could apply to lighting. Peak emission with GE's successful KSF phosphor was ideal for displays at 630 nm. GE is currently developing an alternative red phosphor for the lighting industry with a peak emission of 610 nm, which is more ideal for lighting applications. Using this narrow band red yields an efficacy improvement of over 10%. Murphy highlighted some trends with new architectures beyond on-chip configurations. These included magenta LED configurations, remote part configurations (including remote phosphor film with mini-LED arrays) and micro-LED configurations. He acknowledged that new architectures using downconverters based on mini-LEDs and micro-LEDs are being commercialized. However, he recommended further government investment to help build the ecosystem and ensure a significant U.S. presence in this market.

Jon Schuller, University of California, Santa Barbara: GaN Light Emitting Quantum Well Metasurfaces

Jon Schuller, Professor at UC Santa Barbara, began by providing a background on metasurfaces and their application to lighting. Metasurfaces are phased array optics that can serve as passive optical elements. These nanopillars, which are sub-wavelength in diameter and spacing, can have variations to allow normal incidence light beam and direct the emission at a critical angle. They also can reduce trapped light at the critical angle and provide better photon extraction. An experiment showed that there was a 100x improvement in photoluminescence EQE from strain relaxation and better photon extraction. Schuller described examples of sophisticated metasurface patterns such as a quadratic face profile metalens that creates focused light beams.

Lastly, he discussed metasurface LED designs that his team has explored. Preliminary data on metasurface LEDs shows directed emission with little to no reduction in quantum efficiency.

John Epler, Lumileds: Direct Red Development: Improved AlInGaP VTF and Novel Flip Chip Architecture

John Epler, Device Engineer at Lumileds, described two approaches for direct-red based systems to compete for white lighting: 1) a Red-Green-Blue-Amber (RGBA) LED approach with luminous efficacy near phosphor coated (PC) -white LEDs when at low drive conditions and 2) a PC-mint with direct red, which he described as a practical near-term solution with efficacies that already exceed PC-white luminous efficacy. Most high luminous efficacy LEDs are now vertical thin film (VTF) architecture. Epler described some of the challenges with existing VTF, including non-optimal current spreading and the blockage and absorption of light. Epitaxy, N-metals, and P-metals cause optical losses of roughly 16.2%, 21.6%, and 9.5% respectively. Lumileds is currently developing an upgraded VTF architecture to address these challenges. Measurements have shown a significant improvement in power conversion efficiency (PCE) and luminous efficacy for a test device at 615 nm at 45° C. Epler concluded by stating that to improve beyond this performance would require flip chip architecture.

Kirstin Alberi, National Renewable Energy Laboratory: Red and Amber LEDs, Leveraging Multijunction PV Performance Improvements

Kirstin Alberi, Director of the Materials Science Center at the National Renewable Energy Laboratory (NREL), discussed improvements from III-V solar cell research that NREL has made in arsenide and phosphide multijunction solar cells. Multijunction solar cells have been around for a while but continue to be improved. NREL set the world record for a 3 junction solar cell at 39% efficiency under one sun. This was achieved by 1) modified TJ to improve the top InGaP junction, 2) using strained multiple quantum wells (MQW) for the middle junction, and 3) continuing to improve metamorphic growth for bottom junction. She described how some of these could be applicable to phosphide-based red and amber LEDs. Multijunction LEDs can enable low current operation to reduce efficiency losses in amber LEDs, while the addition of a TJ to single junction GaInP and annealing leads to improved performance through a reduction in point defects. Alberi concluded by describing a dynamic hydride vapor phase epitaxy for III-V systems, which could help lower cost through faster growth.

Ilan Jen-La Plante, Nanosys: Quantum Dot Downconverters for Next-Generation Solid-State Lighting, Challenges and Opportunities

Ilan Jen-La Plante, Senior Staff Scientist at Nanosys, talked about the challenges and opportunities of indium phosphide based or heavy metal free QD downconverters. Narrow red downconverters of any kind can increase luminous efficacy by 5-15% over PC LEDs with broad red emitters. In particular, QD offers flexibility in emission wavelength. One can tune correlated color temperature (CCT) and color rendering index (CRI) easily. Existing commercial products with cadmium selenide based QDs already show energy savings, but require heavy metals (Cd) that are regulated. Nanosys has studied the InP material system to build a fundamental model of power losses as function of flux and temperature. Using their operational model for power loss, the team found that permanent power loss is due to oxidation damage to the organic ligand shell and the inorganic composition. Despite current improvements in power retention, a 3 order of magnitude improvement is required to reach the industry standard of 100,000 hours for L70. Jen-La Plante noted that InP based QD stability under aerobic conditions also needs to be investigated. Nanosys is currently working on: 1) minimizing oxidation by limiting auger hole excitation, 2) continuing to develop novel organic and inorganic shells with a focus on how they interact with the surrounding matrix, and 3) developing encapsulation techniques to provide a gas barrier at QD level. She concluded by noting that one near term application could be to use remote phosphors for diffuse lighting.

Emory Chan, Molecular Foundry: Automated Discovery of Nanophosphors for Solid State Lighting Guided by Machine Learning

Emory Chan, Staff Scientist at The Molecular Foundry, explained the inherent advantages of QD, such as narrow emission and high luminous efficacy while maintaining high CRI. However, the challenge with QD

synthesis is that every layer in the nanoparticle needs to be optimized, with up to millions of combinations of precursors, and multiple variables to optimize (reaction, temperature, time, etc.). This layer-by-layer synthesis is arduous. Furthermore, it is difficult to characterize these structures since they are in the sub-nanometer range. Molecular Foundry combines automation with machine learning by using robots that determine and synthesize quantum dots. Chan explained examples of this approach including 1) liquid handling / lab automation robots, 2) 3D printing robots and incorporating spectral analysis to optimize nanoparticles in real time, and 3) microfluidics. He stated the ideal solution is an autonomous workflow: a closed loop where the machine learning algorithm recommends new particles to synthesize, and the automated robot creates it and characterizes it. This approach allows for rapid convergence on ideal particles. He said that investment in this approach could provide applications for the SSL industry. Potential research topics include multi-objective optimization, developing proxy tests for LED stability, inferring reaction networks, and high-throughput characterizations.

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