DOE Joint Solid State Lighting Roundtables on Science Challenges

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DISCLAIMER

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1. Introduction

Recognizing the potential for coordinated (R&D) efforts, The Department of Energy (DOE) Office of Basic Energy Sciences (BES) and the DOE Energy Efficiency and Renewable Energy (EERE) Solid-State Lighting (SSL) program convened a roundtable discussion among leading experts in LED technology to consider opportunities for further advancement of SSL technology through coordinated R&D actions. The meeting was held on October 7, 2014 at the offices of Navigant Consulting, Inc. in Washington D.C. This report is a summary of the findings from this meeting.

The Basic Energy Sciences (BES) program supports fundamental research to understand, predict, and ultimately control matter and energy at the electronic, atomic, and molecular levels in order to provide the foundations for new energy technologies. Advances in the fundamental understanding of materials may ultimately result revolutionary advances for many energy relevant technologies. The EERE SSL program, on the other hand, has a specific mission to support work that will advance the technology into useful, marketable general illumination products that can produce significant energy savings. EERE SSL funded R&D covers a broad range of activities from applied research in Core Technologies, through Product Development, to Manufacturing R&D. Generally, the R&D projects funded under EERE are on a shorter time scale than those in BES, and there are industry relevant milestones and deliverables to ensure continuous progress.

The DOE BES and EERE SSL Program have a history of working together to explore the basic science needs for SSL with the objective of continuing to advance scientific understanding to support potential technology advances in terms of efficiency, cost, and lighting performance. The first collaborative meeting took place in May 2006 and a second took place in October 2011. Additionally, the SSL program in the Office of Building Technologies of EERE holds annual Roundtable meetings independent of BES to identify areas of critical SSL research needs and to advise DOE on Workshop content and future R&D priorities.

In addition to coordinating R&D efforts, the meeting served to promote discussion on critical applied research priorities for future EERE SSL funding and help identify discussion topics for the January DOE SSL R&D workshop. The roundtable format provided an opportunity for LED experts across the research spectrum to exchange ideas and explore collaborative research concepts. Participants included 14 invited experts in LED-relevant science and technology disciplines drawn from academia, National Laboratories, and industry. They included BES funded researchers, EERE SSL funded researchers, and a few non-DOE funded researchers.

1.1 Objectives and Process

The meeting began with an overview of past EERE and BES collaboration and the EERE Solid State Lighting program. The following four objectives for the roundtable meeting were outlined in the introduction:

1. Identify critical basic and applied research needs for the continued advancement of SSL

2. Foster collaboration among basic science, applied science, and industry researchers
3. Maintain coordination between BES and EERE SSL activities and researchers
4. Gather input and suggestions for the upcoming 2015 SSL R&D Workshop to be held January 27-29, 2015 in San Francisco, California.

In order to gather the input required to meet these objectives each participant was invited to give a 10 minute “Soapbox” presentation describing what they believed to be the critical R&D challenges for SSL. A brief outline of each presentation is found in Appendix A. The presentations were followed by an open discussion of the key challenges. During the discussion, participants were also asked for suggestions on how to enhance collaboration and ideas for the upcoming SSL R&D Workshop.

1.2 Key Conclusions

The meeting format encouraged each of the attendees to participate and present their perspectives on the critical R&D challenges. The discussions that followed the soapbox presentations offered valuable insights into a range of research topics that could advance SSL technology; however, there were a few recurring themes that arose during these discussions that participants felt might provide some fundamental understanding of underlying processes that could lead to significant breakthroughs in SSL performance. These themes are as follows and are outlined in more detail in Section 2:

- Improved down-converters (phosphors and quantum dots)
- Improved materials discovery methods (Materials by Design)
- Efficient and stable red and green direct emitters
- New substrates
- Improved characterization and analysis techniques
- Novel device architectures

Appendix B includes a cross-linking between these critical R&D challenges and existing tasks within the DOE SSL Multi Year program Plan (MYPP).

Additionally, participants expressed concern with the current structure of the SSL program and funding opportunity announcement (FOA) process, where the focus is placed on reaching numerical goals and metrics rather than developing an understanding of the underlying mechanisms. This encourages a “shot gun” approach: trying new materials and structures and hoping to see breakthrough performance without delving into the underlying physics of the materials. It was suggested that DOE should also support projects with the goal of understanding underlying mechanisms associated with the critical R&D challenges, such as the mechanisms of high indium incorporation, the semiconductor physics behind droop, impact of defect structure on carrier dynamics (e.g. droop, non-radiative pathways), and degradation mechanisms for down converters. Focusing on these more fundamental issues could lead to a deeper understanding of the issues which could in turn ultimately lead to improved SSL performance.

2. LED Critical R&D Challenges

2.1 Down-Converters

Developing new and improved down-converters, namely phosphors and quantum dots (QDs), was clearly identified as a science research topic that could have a significant impact on improving SSL technology. For both phosphors and QDs, there should be a focus on understanding the impact of materials choices and designs on efficiency, spectral characteristics, wavelength tunability, and line width control. For practical applications it will be necessary to embed the down-converters in a matrix material such as silicone, and research is required to understand and control the complex interactions (e.g. chemical, optical, thermal, and degradation interactions) between the down converter and matrix. There also needs to be work on improving our understanding of degradation mechanisms of down-converter materials with respect to temperature, flux, humidity, and the combined impacts of these effects.

2.1.1 Phosphors

Narrowing the emission of red phosphors can reduce the amount of light emitted outside of the visible spectrum and enable higher efficacies for white LED packages. Similarly, narrower phosphors in other spectral regions could enable higher efficacies. Developing phosphor systems that offered spectral tuning would also provide improved control over the trade-off between CRI (color rendering index), CCT (correlated color temperature), and efficacy. Thermal quenching in phosphors leads to reduced efficiency at elevated temperatures and therefore color shift of the emitted white light from a blue-LED pumped source. Phosphors with reduced thermal quenching can improve color stability and simplify the design constraints for higher power LED and laser-based lighting by allowing for higher operational temperatures. However, thermal instability in phosphors is currently poorly understood, and more fundamental research is needed.

2.1.2 Quantum Dots (QDs)

Further work is required to understand stability issues associated with QDs such as thermal quenching, photobleaching, blinking, and environmental stability. For example, there are at least two mechanisms for blinking; charge (Auger) related and hot carrier related. Interactions between the shell and core of quantum dots also needs to be studied. In this respect, current research on “giant” QDs, which are much larger than traditional quantum dots due to increased thickness of the shell, might provide some answers. There is also a push for cadmium-free QDs due to concerns over toxicity, but there is a lot more research and development required to make stable, efficient QDs without cadmium. Nevertheless, alternative cadmium-free solutions such as Indium Phosphide (InP)-based QDs should continue to be explored. For QDs, another big issue is matrix compatibility. The QDs and the matrix material (often silicone) need to be studied together in order to understand the reliability of the system.

2.2 Improved Materials Discovery Methods (Materials by Design)

The concept of exploring and developing new materials for LED lighting systems including new phosphors and LED emitter materials through computational processes was brought up at the meeting. For example, in the case of phosphors, if the fundamental physical mechanisms of the light emission processes could be understood with respect to the material properties, then it is conceivable that materials could be modeled and engineered to achieve the desired light emission properties. This approach could be continually refined as our understanding of the relationships between different materials combinations and their light emission properties improved. Other materials in the LED package, including the LED emitter itself or the thermal and optical materials could also, conceivably, be explored and designed through computational processes.

2.3 Red and Green Direct Emitters

Phosphor converted LEDs are fundamentally limited by Stokes loss and losses in the phosphor conversion process. The development of more efficient direct emitting red and green sources eliminates Stokes loss and offers an alternative route to the efficacy target of 250 lm/W and beyond. Not only do these emitters need to be efficient, in terms of internal quantum efficiency, but they should also have stable emission with respect to temperature, in order minimize color variation between the different emitters at different temperatures.

There are fundamental materials problems that limit the performance for AlInGaP red and green LEDs. The efficiency of AlInGaP LEDs drops as the wavelength moves from red to green due to the energy band line-up (indirect gap). Also, the efficiency of the AlInGaP materials system drops rapidly with increasing temperatures compared to the temperature stable InGaN materials system. An all InGaN based solution has the potential for improved temperature stability if materials issues associated with the high indium compositions required for green and red emission can be overcome. Research focused on developing an efficient InGaN red LED could have the benefit of leading to an efficient InGaN green LED (which uses lower indium compositions) in the process.

2.4 New Substrates

New substrates can potentially enable improvements in LEDs, particularly for red and green emitters. For example, a novel Scandium Aluminum Magnesium Oxide ($ScAlMgO_4$ or SCAM) substrate is lattice matched to InGaN at compositions suitable for green wavelength emission (~ 17% indium). New substrates with tailored lattice constants might help alleviate strain at longer wavelengths, which could lead to more efficient long wavelength InGaN LED emitters.

There is also continued interest in pursuing bulk GaN substrates, which remain expensive but have the potential to reduce dislocation densities, polarization fields, and droop in LEDs, and to reduce epitaxy costs.

2.5 Improved Characterization and Analysis Techniques

The development and application of new and novel characterization and analysis techniques may provide a path to solving many challenging materials issues. Characterization techniques such as deep level optical spectroscopy (DLOS), synchrotron x-ray diffraction, and Electron Emission Spectroscopy (to measure Auger generated hot carriers) can be used to analyze the materials properties of LEDs, thus leading to greater understanding of challenging issues including; (i) the identification and elimination of point defects and other non-radiative pathways, (ii) reducing strain in high In composition LED active regions, and (iii) monitoring Auger recombination processes which lead to current density droop. In addition, advanced characterization techniques can help improve our understanding of degradation mechanisms in down-converters (phosphors and QDs). This improved understanding of materials characteristics should lead to the development of predictive modeling and verification of these models through testing on state-of-the-art materials.

2.6 Novel Device Architectures

New device architectures can be explored to mitigate performance barriers that exist in more conventional LED architectures or offer new functionality. Below are a few examples that were highlighted.

2.6.1 Lasers

Blue laser diodes have a distinct efficiency advantage over blue LEDs at very high current densities. For laser diodes, current droop is eliminated when lasing occurs. This enables high flux density and higher wall-plug efficiencies than LEDs at very high current density operation. In addition to a higher efficiency, laser diodes can allow for more effective use of emitted photons due to directional beam characteristics, the potential for higher luminous efficacies in color-mixed white sources due to narrow linewidths, and possibly smaller form factors for optical systems and unique luminaire designs. Laser-based white lighting systems operating at high photon fluxes have already been demonstrated for commercial automotive applications, but these are expensive and complicated systems. High-power laser diodes have a somewhat more complicated epitaxy process, higher substrate cost (compared to most LEDs), a shorter device lifetime, and, in general, lower efficiency than LEDs. Laser lighting could be used in some specialty, high etendue applications, but in order to make a broader impact on the lighting market these issues need to be overcome. In addition, reducing the threshold current density of laser diodes could enable their benefits at lower current densities making them more effective for use in SSL.

2.6.2 Multi-Junction LEDs

Novel emitter architectures based on the use of tunnel junctions within the LED heterostructure have recently been demonstrated. Tunnel junctions can improve hole injection into the active region and enable the stacking of multiple pn-junction active regions. The multi-junction LED structure operates in a high-voltage/low-current regime and can produce the same light output as a conventional single junction device at a lower current density, thus minimizing the effects of current density droop. Another benefit of the lower operating current is less joule heating which could simplify thermal management issues.

2.6.3 EL QD LEDs

Electroluminescent (EL) QD LEDs are another possible solution to the requirement for improved high efficiency narrowband red emitters. While QDs are typically applied as photoluminescent (PL) elements (down-converters) within the LED package, they can also be directly electrically pumped. In order to efficiently electrically pump QDs they need to have specific electrical properties and efficient charge injection layers. Red EL QD LEDs have demonstrated 18% external quantum efficiency (EQE) at 620 nm.

3. Collaboration and Coordination

One of the goals of the BES/EERE roundtable meeting was to identify scientific areas for collaboration and to find ways to enhance the transfer of basic science discoveries to product development programs in EERE. Accordingly, a portion of the roundtable was set aside to discuss collaborations among BES and EERE efforts. Participants agreed that collaboration would be advantageous for advancing solid state lighting technologies.

Everyone was in agreement that if research is to be performed to further our understanding of complex mechanisms, it should be done with the best quality material so that other materials issues do not obscure the results; however, National Laboratories and universities often do not have access to the commercial materials used in industry, thus presenting a clear opportunity for collaboration. The reasons for the current lack of collaboration range from cost sharing restrictions (for National Laboratory participation in EERE FOAs) to sensitivities surrounding intellectual property. Participants agreed that if DOE funding opportunity announcements specifically asked for collaboration, they would be encouraged to do so in order to gain an advantage in the competitive process.

During this discussion, participants representing the National Laboratories identified another avenue for collaboration: university and industry groups could take advantage of the user facilities at DOE National Laboratories.[1] Researchers can use the facilities for either research that will be published in the open literature or confidential research, under non-proprietary and proprietary user agreements respectively. Access to the facility requires the submission of a user proposal, which is reviewed, typically by an external group of peers, to determine priorities for access to instrument time at the user facility. Approved users have access to the capabilities as well as assistance of expert user scientists. For non-proprietary research, use of the DOE facility is free. Proprietary research requires full cost recovery; PIs should contact the facility directly to get cost estimates for the type of research to be done. Under these agreements, researchers may collaborate with National Laboratory scientists or independently use the facilities.

4. Suggestions for the DOE SSL R&D Workshop

The 2015 SSL R&D Workshop, which is to be held January 27-29, 2015 in San Francisco, CA, offers another opportunity to continue the discussion on critical R&D challenges. Another goal of the BES/EERE roundtable meeting was therefore to gather input and suggestions for suitable topics and speakers, and ideas for panel discussions. Participants suggested the following:

- It would be helpful to have a presentation on how DOE user facilities could be impactful and how to use them. Particularly if you can find the right speaker to give an example of a success story.
- There is a disconnect between the people who make LED packages and the luminaire manufacturers who use them. It would be helpful for each to hear about the expectations and challenges of the other.
- Set up topic tables by critical topic area rather than based on the existing task structure so as not to limit feedback.
- Key topics for presentations and panels include: red emitters, down-converters, reliability, and color control.

[1] For more information please see: http://science.energy.gov/bes/suf/user-facilities/ and http://energy.gov/gc/access-high-technology-user-facilities-doe-national-laboratories

Appendix A: Attendee Presentation Summaries

Christian Wetzel, Rensselaer Polytechnic Institute

Epitaxial Materials Developments for Direct Emitters

In order to make all LEDs efficient and maximize white light efficacy, we need to work on efficiencies in the green gap. Efficient LEDs are currently available for blue emission but not yet for longer wavelengths. Consequently the industry relies on phosphors to get light that appears white. Direct LEDs are what we need, but we aren't able to do that efficiently with green yet. For longer wavelengths, defects matter more than they do for blue. One answer to relieve the strain and resulting defects may be the use of a strain matched substrate. There is a new possibility in this respect, a substrate called SCAM ($ScAlMgO_4$), which has better lattice matching for the high indium compositions required for green emission wavelengths.

Device motivated core research needs to include work on the growth of high indium composition alloys, polarization control in nonpolar cubic growth, mismatch strain control during growth, and alternative substrate development.

Andy Armstrong, Sandia National Laboratories

Innovative materials characterization, synthesis, and active region design to overcome the green gap and droop

Point defects in materials operating in the green, yellow, and red regions are poorly understood and controlled. There is a big increase in point defects when we move from blue to green, and this is critical. We need to know what the atomic sources of the defects are. Deep level optical spectroscopy (DLOS) can be used on LED quantum wells to probe mid-gap non-radiative recombination centers and quantify the quantum well defect energy level and density. DLOS has nanoscale depth resolution which allows it to probe quantum wells individually and is non-destructive. The use of new characterization techniques to understand the materials challenges may provide a path to solving many challenging issues in LED emitter materials. There is also a need for fundamental materials research on LED quantum wells to understand and mitigate the impact of defects on radiative efficiency, especially at long wavelengths. Bulk GaN substrate development is not a panacea for eliminating defects. Extended defects may be reduced through the use of bulk GaN substrates, but what is the impact on point defects? There is a very strong depth dependence of quantum well defects, and the p-region has fewer defects than the n-region. The point defects in multiple quantum well devices depend on active region design as well as growth conditions and indium mole fraction. Toshiba has shown that you can get better results in green efficiency with InGaN quantum wells capped with aluminum gallium nitride (AlGaN) interlayers. Sandia has reproduced these results and shown that the improvement comes from sharper hetero-interfaces, with the AlGaN enabling the use of higher GaN quantum barrier growth temperatures that improve quantum well efficiency (defect reduction), and create higher polarization-induced electric fields in the quantum wells for longer wavelength emission.

Novel architectures can also lead to improved performance. For example, the Rajan group at Ohio State University is stacking several LED junctions to reduce droop. Less joule heating reduces luminaire cost, adding a cost benefit in addition to efficiency benefits.

Paul Fuoss, Argonne National Laboratory

Synchrotron X-Ray Studies of the Synthesis and Processing of Wide Bandgap Semiconductors

ANL has a group that is focused on the use of advanced photon sources to study the synthesis of a variety of materials including wide band gap semiconductors. The equipment enables real-time x-ray diffraction studies to be performed during the metal organic chemical vapor deposition (MOCVD) growth of GaN. MOCVD growth of GaN is complex; surface catalysis plays an important role, step and dislocation dynamics lead to non-planar growth, and growth on different planes is very different (e.g. c-plane growth is hexagonal, m-plane growth is not). It is important to talk about strain at the nanoscale rather than the macroscale. Stress and stress relaxation in GaN/InGaN structures play a crucial role in the synthesis, processing and performance of LEDs. Syncrotron X-ray studies have been performed on such structures to image strain at the nanoscale. The coherent x-ray beam produces complex patterns referred to as “speckle”. From these patterns it is possible to reconstruct phase information which is directly related to strain in the device. This enables 3-D reconstructions of the strain state from 2-D diffraction data more efficiently and with a simpler experimentation than previously possible. Opportunities for Synchrotron X-ray studies include a systematic study of growth on different crystal faces revealing underlying diffusion and attachment mechanisms, gaining nanoscale information (e.g. strain) during device processing and operation, imaging studies of the formation of islands and defects in situ during growth, and examination of the evolution of strain and composition around defects during operation.

Bill Tumas, National Renewable Energy Laboratory

A Photovoltaic materials science perspective on Solid-State Lighting research needs and directions

There has been a lot of improvement in SSL since 2006, which begs the question where are the asymptotes and when will we see them? We can’t think of light as the same old photons, we need new things. Next generation concepts for color mixing include metamorphic growth for higher efficiency amber/green inorganic LED materials and new device architectures. Quantum dots are another important topic, but they have their challenges. Advantages to using quantum dots include inexpensive manufacturing through scalable solution growth and processing, material tunability, added degrees of freedom in materials and new device design using nanoparticles as building blocks, and new physics of multiple carrier generation, plasmonics, nanoscale charge-transfer and high PL quantum yields. Challenges include producing narrow line widths, reducing size disparity, achieving high PL quantum yields, developing scalable synthesis methods, developing solution-phase device fabrication, and achieving stability under typical operating conditions.

For OLEDs, we need to get improved blue emission, and to achieve this, the focus needs to be on layers other than the active layer. For example, important science is occurring between the electrode and active layer. We need efficient utilization of triplets in OLEDs, and are interested in the thermal conversion of triplet to singlet. We are interested in controlling triplet and singlet energies of the host material to facilitate efficient energy transfer in the blues. There is a need for integrated materials development, a synergistic approach to combining theory and modeling for targeted structures, high throughput and targeted synthesis, and full material and device characterization.

Specific R&D needs for interfacial materials and contacts for LEDs/OLEDs include:

a) better understanding of interfacial electronics and stability; materials and process based approaches to optical management
b) examination of new material combinations and process approaches to enable multifunctional contacts
c) charge selective contacts, e.g. p-type nickel oxide (NiO_x) and deep work function n-type Molybdenum Oxide (MoO_x), which can enhance OLED performance
d) improved understanding of device interfacial carrier and photon effects to create design targets for material properties to enhance performance

Russell Dupuis, Georgia Institute of Technology

<u>A chicken in every pot and two UV lasers in every garage –and more in the house!</u>

For the future development of SSL we need to ask, "What is important?". Current technologies are not the end, and LEDs still have work to do. It has been shown that InGaN blue laser diodes have a distinct advantage over blue LEDs at high current densities (and high photon densities) with the prospect of higher wall-plug efficiencies due to reduced "droop". Lasers also offer more effective use of emitted photons due to directional beam characteristics, higher luminous efficacies due to narrow linewidths, and smaller form factors (potentially enabling unique luminaire designs). High-efficiency white lighting systems operating at high photon fluxes have already been demonstrated for commercial automotive applications, but in these platforms cost is no object.

As primary sources for lighting, high power LEDs have a somewhat "simpler" epitaxy process, but only on sapphire and silicon carbide (SiC) (it is more complicated for low-cost substrates e.g. silicon). Also, LEDs are only efficient at low current densities before droop takes over, and complicated fabrication processes are required to create high extraction efficiency, thin film LEDs. High-power laser diodes have a somewhat more complicated epitaxy process, higher substrate cost (compared to most LEDs), and a shorter device lifetime, but they can have more devices per unit area, and the directional beam enables additional system efficiencies and novel luminaire designs. For current state of the art ultraviolet, blue, and green laser diodes it has been demonstrated that high external quantum efficiency is possible in a wide wavelength range from 400 to 480 nm, and laser diodes are available commercially for 400-405, 450, 468-478 and 525 nm wavelength ranges. It should also be possible to create high efficiency ultraviolet, UV-A laser diodes at wavelengths around 380-400 nm.

Five year goals for laser diode development should include:

a) improve MOCVD epitaxy for AlInGaN-based UV-A emitters
b) evaluate AlInGaN quaternaries for UV emitters
c) optimize active region and device designs for more effective carrier injection
d) reduce resistive losses in wider-bandgap AlGaN cladding layers
e) study potential alternative high-power laser device structures
f) evaluate nonpolar and semipolar UV-A lasers
g) evaluate improved substrate technologies
h) develop facet coatings for improved device reliability
i) study lifetime limiting processes.

Nathan Gardner, Glo

<u>High Efficiency Temperature Stable Red…and Green: Emitter Materials Research</u>

There is a need for high efficiency temperature stable red LED, which would lead to efficient green (needed as well). RGB+ lighting is really the path to 250 lm/W warm white light since phosphors and down converters will always have Stokes loss. Down converters work if you have no other option, but if you can create photons you want directly at colors you want, you will have better efficiency. Also, RGBA (red, green, blue, amber) direct emitters give the additional functionality of color tunability. If you can get to 250 lm/W you can really compete with fluorescents and can make incandescent replacement lamps even cheaper because there can be less thermal management.

RGB, RGBA, and hybrid approaches need a red direct emitter with higher power conversion efficiency and good temperature stability. There is a fundamental energy band problem for conventional AlInGaP-based red LEDs which limits the power conversion efficiency (PCE) at shorter wavelengths, and seriously impacts temperature stability. This behavior is associated with a direct-indirect band crossover effect but this doesn't occur in InGaN-based alloys so they could hypothetically produce an efficient and temperature stable red emitter. In practice the performance of longer wavelength green and red emitting materials is limited by other factors that we don't fully understand. For example green PCE has been stuck at 22% for years now. Is it a fundamental material issue that we have not recognized? Is the issue point defects, and if so what are they? If we knew these answers we could control them, but right now fundamental emitter questions are expensive to answer because they are investigated by empirical growth studies. These burn time and money because of MOCVD reactor and material requirements, with more direction we could help reduce expense. Investigating the basic technological problems will fundamentally alter the cost and performance trajectory of SSL. Such studies are high risk and high reward (keep US companies competitive).

Note: The red LED power conversion efficiency of 44% listed in the 2014 MYPP Table 3.4 is for room temperature operation, at 80°C the efficiency would drop to 22%.

James Ibbetson, Cree

<u>Aligning New Technology with Lighting Operating Conditions</u>

Cree will employ any and all technological advances to improve LED package performance and rapidly bring these advances to market. Today, LED package performance at 25°C is near the 2013 goal at a current density of 0.35 A/mm^2; however, we need to get away from quoting performance at room temperature since 105°C is more relevant for real luminaires and lamps. Historically, we plot luminous efficacy versus the current (or current density), and the choice of operating at 0.35 A/mm^2 is arbitrary. A push to higher efficacy through reductions in current density drives up the cost, and instead the market is pushing for higher current densities because that is cheaper. Therefore we need to develop approaches to achieve higher efficacy at higher current densities.

The DOE targets 220 lm/W by 2020, but we have a long way to go to get there, and 6 years is not a long time. To get there we will need to do a lot of different things that can each give small gains that add up, but there are fundamentally two different approaches:

a) emphasize operation at low current densities and temperatures, minimizing the impact of droop, reducing the demands on phosphors etc. (Type 1)

b) emphasize operation at high current densities and temperatures, minimizing semiconductor material usage but placing increasing demands on droop reduction and more stable phosphors (Type 2)

Type 2 gains are preferred for faster and wider SSL adoption because we will see real energy savings sooner rather than imagined savings later, and set new technology on its own cost/volume trajectory. Very low cost epitaxy will allow the Type 1 approach to succeed, but the ability to address a Type 2 approach offers us a high risk, high reward strategy that could set us apart. If we want to be competitive with China we need to embrace the Type 2 approach. Hence we should focus limited resources on technologies that in 3 to 5 years can help improve operation at high flux density and temperature. For example, reducing droop by characterizing Auger recombination mechanisms as a function of junction temperature, developing a new narrow band red phosphor that is efficient and reliable at 105°C and 0.5 W/mm^2, and developing high reflectivity plastic packages with long term stability at 105°C.

Mike Krames, Soraa

Core Research Challenges for Breakthrough LPW

The Nobel Prize was given specifically for blue LEDs, and in fact nearly all of the gains have been in the blue and the violet spectrum resulting in fantastic devices that drive the industry today. However there is still a green gap, and much work remains to be done. Gains targeted in the 2014 MYPP will be difficult to get to with a phosphor only system. A breakthrough is required, and it needs to be something significant that we haven't really seen before.

Auger recombination and droop are still a problem, especially when you get to wideband gap materials where recombination channels get more complicated. We'd like to operate at higher current densities but for that we pay 10, 15, or even 20% efficiency penalties. We need more money going into understanding recombination pathways instead of avoiding the problem by simply lowering the current density. Lasers are a possibility if you can achieve a low threshold current density, since the carrier density is fixed at threshold and this clamps Auger recombination which effectively eliminates any further increase in droop.

Down-converters are another area that requires improvement in terms of color stability and reliability, especially at high temperatures. We need narrow red emitters, "line-emitting" phosphors, or non-phosphor down-converters. QDs require developments in terms of temperature management and flux density so we can realize effective "on-chip" performance. We should also look into cadmium-free QDs. QD issues are solvable but require more investment.

We should remove outdated tasks from the DOE focus such as:

a) light extraction approaches, because we know where the gaps are and its being worked on
b) electronics reliability research, because unless it is something novel it is well understood
c) manufacturing simulation.

Instead we should focus on "big impact" tasks such as understanding materials systems and Auger recombination. These need a lot of growth runs, detailed analysis time and the involvement of different skill groups which won't come cheap, so there needs to be funding there. It would be helpful for proposal writers if we were to remove outdated tasks not just prioritize all tasks.

Jim Neff, Philips Lumileds

LED Color Conversion for 250 lm/W

It will take a lot of research to get high efficacy white LED architectures at 3000K with 80 or 90 CRI and 250 lm/W. We should focus on phosphor-converted (pc) solutions because those technologies are able to be delivered by 2020, at which point the lighting revolution may be nearly over. Efficiency losses for pc-LEDs come from blue pump LED internal quantum efficiency (IQE) droop and also phosphor conversion efficiency, which is where we think there are improvements to be made. For color mixed LEDs, losses come from weighted power conversion. We need practical architectures and I think this means pc-LEDs. Color mixing will have its place but it won't be in the low cost general light bulb at Home Depot and it won't allow us to replace the troffer. Consumer expectation is that LEDs will fix every problem lighting has ever had; it can't just be as good but it must be better. For example, people are even asking for 90 CRI streetlights.

Pc-LEDs can make it to 250 lm/W with a CRI Ra > 80 and unconstrained R9, but it is ambitious. The product of wall plug efficiency, down conversion quantum efficiency, and package efficiency must be >0.75. For CRI Ra > 90 and R9 > 50, 250 lm/W is not possible and 225 lm/W is a better target (at 85°C), but we need narrow red and narrow green emitters with good quantum efficiency to get there. We also need robust packaging and silicone reliability, and high efficiency blue pump LEDs operating at high current and high temperature.

Jennifer Hollingsworth, Los Alamos National Laboratory

Next-Generation Giant Quantum Dots: Solving the Solid-State Performance Conundrum

Our work is BES funded, but it is 'use inspired' science with a focus on SSL. We are working on giant QDs (g-QDs) that have diameters of 15-20 nm which is much larger than traditional quantum dots. The structure of the g-QDs, a small core with a thick outer shell, gives novel functionality. Emission occurs exclusively from the core, and absorption occurs from the "antenna" shell which results in large effective Stokes shift and minimal self-reabsorption. The g-QDs do not photobleach, are non-blinking, resist saturation at high flux densities (i.e. non-radiative processes such as Auger recombination are suppressed, and efficient emission results from charged and multiexciton states), and emission is largely independent of surface ligands. The shell thickness is why the QDs are called "giant", but we found that core size is also very important particularly with respect to blinking. Core size and shell thickness are together 'tuning parameters' for either high or low biexciton emission efficiencies coupled with suppressed blinking.

We found that g-QDs outperform standard QDs in direct-injection devices; with EQE greater than 10x higher, luminance more than 1000x the standard (2000 Cd/m^2), and down-conversion efficiencies of up to 88%. A new automated reactor system is being employed to help meet the "scale-up" challenge for these nanomaterials. Computer controlled synthesis allows for automated precursor delivery, material sampling, and in-situ diagnostics. The system also facilitates quasi-combinatorial materials exploration which will help us more rapidly discover new g-QDs, assess and address temperature quenching, and evaluate their reproducibility and suitability for subsequent scale up.

Research needs include improving the green QD performance, enhancing the color tunability of g-QDs, and improving the reproducibility of the synthesis process.

Jeffrey Pietryga, Los Alamos National Laboratory

Nanoscale Engineering of Quantum Dots for SSL: QD Phosphors and LEDs

BES is interested in the fundamental interactions between light and QDs. Several advantages motivate the use of QDs as down-converting phosphors including bright photoluminescence, selectable energy and bandwidth over the whole visible spectrum, low cost and scalable manufacturing, and the elimination of "critical" rare earth elements. However development is still needed to retain efficiency at elevated operating temperatures, to achieve longer lifetimes, and retain full compatibility with compositing techniques. For example, silicone is a particular favorite matrix material used to apply down-converters to LEDs, but silicones formulated with a platinum catalyst can be deleterious over time due to complex interactions with QDs. We need to co-develop QDs and the matrix material.

In an alternative application (a solar photo concentrator), we have found a new way to embed QDs in plexiglass (PMMA) while maintaining 95% quantum yield for a solar window.

Another interesting approach is to use QDs as the active region in the LED. For QD-LEDs, advantages include potential efficiency gains and reducing costs at large scale, but brightness must be enhanced (through higher efficiency and less droop) and lifetimes improved. Charged QDs produce Auger losses, not because of high carrier density, but because the charge is imbalanced (causing spontaneous electron injection).

There is the potential for cadmium free QDs, and that is important. One option is a copper indium sulfide core with a zinc sulfide shell ($CuInS_2$/ZnS). Retaining QD performance in a practical application and achieving a technologically relevant lifetime will require "hardening" of the QDs using chemical and heat treatment without sacrificing PL efficiency. Co-development of the matrix material and QDs will also be important. Bright long-lived operation at desired color point will require further engineering of reduced Auger charge-resistant QDs, use of advanced spectroscopy to analyze long term failure modes, and reduced organic content (likely to enhance stability). Additionally, reduced use of vacuum processing steps will minimize cost and scalability problems.

Vladamir Bulovic, Massachusetts Institute of Technology

UV-Vis-IR Quantum Dots for SSL

QDs span the visible spectrum, and within the visible spectrum QDs are close to meeting the MYPP's 2020 goal of less than 30 nm full width half maximum (FWHM) (Task A.1.3). QDs used in enhanced LCDs have a FWHM of 50nm, and can get down to 25nm with careful synthesis. These phosphors are only one millimeter away from the backlighting LEDs in the TV and reach temperatures of 70°C. QD synthesis can be scaled economically, but managing waste is important. With a single-step synthesis process, CdSe/CdS QDs are estimated to cost around \$61/gm or \$4/m^2 compared to Ir(ppy)$_2$, the emitter material used in OLEDs which costs \$658/gm. A state of the art EL QD-LED with a brightness of 10,000 cd/m^2 at 5 volts has an EQE of 18% and an IQE of about 90%, which is satisfactory. Beyond the visible spectrum there are infrared LED applications such as telecommunications, bio sensing and spectroscopy, bio-medical imaging, and military technologies. Other applications include sleep, food heating, UV water treatment/pasteurization, phototherapy (e.g. treating neonatal jaundice with blue light), and UV curing.

Jakoah Brgoch, University of Houston

Advancing Phosphor Science: Targeting high-efficiency, thermally robust phosphors

Work is being performed on novel phosphor development, advancing phosphor science by targeting high-efficiency, thermally robust bulk phosphors. We need a better approach to the identification of suitable candidates for efficient inorganic phosphors with improved color rendition and for selecting efficient phosphors at specific target colors.

High efficiency phosphors are currently at about 90% down conversion efficiency, but they are selective. To reach the target efficiencies identified in the program we must seek to optimize known phosphors but there is a limit to how much more we can do. So we may need to identify new phosphors instead. In this respect we need a better approach to developing efficient inorganic phosphors with better color rendition. This might mean looking at a material genome and encouraging multifaceted research that combines computation, data-mining, and experiment as opposed to just picking materials and trying them. We also need to develop phosphors that have enhanced thermal quenching properties for high power LED and laser based lighting as the sources are more powerful. Going from room temperature to 150C, there is currently a 10% drop in PL quantum yield, but the 2020 target is to reduce that to 5%. Thermal stability is currently poorly understood and fundamental research in thermal quenching is needed. Models exist to interpret data but not to predict thermally robust emission. Using computation and experiment in tandem will produce models that can be used to target high temperature phosphors, and it should be a focus of R&D efforts.

Jim Murphy, GE Research

Narrow-band Phosphors for SSL

There is a tradeoff between efficacy and CRI. It has been well-known since the 1960's that narrower line-emission enables higher efficacy for the same CRI. This effect is stronger for red spectral regions compared to green or blue because the human visual response falls off rapidly in the deep red. Typical red phosphor material has a QE greater than 90%, but because the FWHM is > 90 nm, a large portion of the spectral emission is at wavelengths greater than 650 nm, which is beyond the eye response, leading to efficacy losses.

There are currently two possible routes for achieving narrow band phosphor red-line emission, QDs and manganese (Mn^{4+}) fluoride based phosphors. For other colors, only QDs currently meet spectral and absorption requirements. The need for new line emitting LED phosphors requires core or basic science programs. Typical RE^{3+} activators have low absorption and require sensitization. Transition metal activators except Mn^{4+} do not meet spectral requirements. Both sets of line emitters based on these activators have slow decay times which lead to intensity saturation. Another challenge is that once you have identified a new material, reliability is not well standardized and difficult to assess. Some understanding exists regarding photo-oxidative processes and hydrolysis, but there is less understanding regarding the relationship between defects, processing, and performance. For instance a small parameter change in these areas can cause huge changes in products at 50,000 hours. Phosphor reliability is tested at 85°C/85% humidity for 150 hours. By working on reliability, the high temperature, high humidity performance can be improved. Different phosphors are currently used for high and mid power LEDs. Reliability improvements would allow for a broader phosphor portfolio in mid power devices.

Appendix B: Linking of Critical R&D Challenges to Existing MYPP Tasks

Critical R&D Challenge	Related MYPP Task
Improved down-converters (phosphors and quantum dots)	A.1.3 Down-converters
Improved materials discovery methods (Materials by Design)	A.1.2 Emitter materials research A.1.3 Down-converters
Efficient and stable red and green direct emitters	A.1.2 Emitter materials research
New substrates	A.1.1 Alternative substrates
Improved characterization and analysis techniques	A.1.1 Alternative substrates A.1.2 Emitter materials research A.2.2 Novel emitter materials and architectures
Novel device architectures	A.2.2 Novel emitter materials and architectures