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Lighting R&D Program: Diffuse Light Sources R&D Meeting

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DOE Lighting R&D Program, "Diffuse Light Sources R&D Meeting," February 2021.

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Comments

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Diffuse Light Sources R&D Meeting Participants

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Tommie Royster	R-Display and Lighting
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Table of Contents

1 Introduction

On October 8, 2020, experts in organic and quantum dot light emitting devices gathered at the invitation of the Department of Energy (DOE) Lighting R&D Program to help identify critical research and development (R&D) topic areas in the field of diffuse light emitting materials. This year, for the first time, the meeting was held virtually due to travel difficulties and concerns related to the ongoing COVID-19 pandemic. The meeting commenced with "soapbox" presentations where each participant was invited to give a short presentation describing the state of understanding of organic light emitting diode (OLED) and quantum dot electroluminescent (QDEL) materials for lighting, gaps in understanding, and R&D suggestions to advance understanding. This was followed by a general discussion of the R&D challenges and barriers to implementation. Following these discussions, the participants were asked to contribute ideas regarding program content for the upcoming R&D Workshop to be held February 1-4, 2021.

The meeting format provided an opportunity for OLED and materials experts in the field to exchange ideas and explore collaborative research concepts. Participants included invited experts in OLED and QDEL lighting-relevant science and technology disciplines drawn from academia and industry. Attending companies include a diffuse lighting panel manufacturer, materials manufacturers, and a computational chemistry group. They included DOE-Lighting R&D-funded researchers as well as non-DOE-funded researchers.

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

1.1 Key Conclusions

The meeting format encouraged each of the attendees to participate and present their perspectives on critical R&D challenges. The discussions that followed the presentations offered a variety of valuable insights regarding diffuse light sources; however, there were some recurring themes that arose during these discussions regarding research areas that could advance the development of source materials, especially for blue emitter systems. These themes are as follows and are outlined in more detail in Section 2:

- Stable and Efficient Blue Emitter Materials – including blue phosphorescence, thermally assisted delayed fluorescence (TADF), hyper-fluorescence (TADF-assisted fluorescence), metal-assisted delayed fluorescence (MADF) and QDEL
- System Solutions and Partnerships – the value of developing all materials in the emitter system congruently and the important role of collaborations in this development
- Alternative Approaches – new ways of solving current OLED materials shortcomings, such as dilution of organic semiconductors to alter layer properties, simplified structures, high entropy hosts, and novel device architectures

2 Critical R&D Topic Areas

2.1 Stable and Efficient Blue Emitter Materials

In order to create high performance diffuse lighting, stable and efficient blue emitters are needed. There are many materials approaches to overcome this decades-long challenge. TADF, phosphorescence, hyper-fluorescence, and metal complex based delayed fluorescence are all being investigated as potential solutions. These approaches all have promise, but none have simultaneously achieved the efficiency and stability needed for the blue component of diffuse OLED or QDEL lighting. Many white OLED (WOLED) lighting devices currently use a hybrid approach comprising red and green (or yellow) phosphorescent emitters with a fluorescent blue emitter because the stability of blue phosphorescent materials is unsatisfactory. However, fluorescent materials fundamentally have insufficient harvesting of triplet excitons, thus resulting in poor efficiency. Current external quantum efficiency (EQE) is only around 10% for fluorescent blue, thus alternatives are desperately needed.

Commonly, phosphorescent approaches have been researched, using a heavy metal-based emitter for high luminescence efficiency. Phosphorescent materials have the advantage of almost 100% internal quantum efficiency (IQE), the spectrum can be tuned by ligand design, and they offer a wider spectrum than fluorescent emitters. However, they tend to require higher voltage and have poor lifetime. TADF has been vigorously investigated in the past 5-10 years as a potential alternative to phosphorescent emitters due to their similar, near unity IQE, wide spectrum, and potential for cost reduction over phosphorescent materials. However, TADF shares the same lifetime shortcomings as phosphorescence and attendees suggested that this approach is appearing to be unsatisfactory for blue. Hyper-fluorescence uses the ~100% exciton harvesting of TADF dopants to generate excitons and the excited energy then transfers to a fluorescent molecule. Four times more light is emitted by the fluorescent in the TADF-assisted approach as compared to conventional fluorescence with the same input energy. Multiple attendees indicated that hyper-fluorescence seems the most promising approach as a deep blue color, a high color purity, and high efficiency which can be achieved at potentially reduced cost. The downsides of this approach are not only lifetime, but also a more complex system which requires a host, TADF assistant dopant, and fluorescent emitter to be developed in concert.

QDEL approaches also struggle with the blue stability and lifetime. Quantum dots are attractive materials for diffuse emitters because they have high IQE, high luminance at low voltage, they emit with an easily tailorable narrow spectrum, and they are solution processable for low cost. EQE of blue is currently around 12.7% and lifetime LT50 (the time it takes until emitted light reaches 50% of the initial output) at 1,000 cd/m^2 is around 350 hours. This technology has great promise, but much work is needed to improve the performance of quantum dot emitters and integrate them into white device structures.

Throughout the discussion, attendees put forth metrics for blue light. The blue light should be deep enough for high color rendering index (CRI), with maximum emission between 457 nm and 470 nm. Blue emitters should also yield a high luminance per current density. Lifetime LT90 (the time it takes until emitted light reaches 90% of the initial output) of 500 hours at 1000 cd/m^2 is required and low materials cost is another key consideration.

2.2 System Solutions & Partnerships

Participants stressed the need for solving materials issues with complete system solutions. For optimal light generation and stability, hosts and dopants must work harmoniously. Device efficiency and stability reflect the quality of the individual components and the system as a whole. Investigations solely focusing on one component of a blue light system have limited value until the work is done to integrate them into a complete system with hosts, emitters, assistant-dopants, charge generation and transport layers that may include blocking layers.

In order to achieve co-development of all molecules for a blue emitter system, strategic partnerships are key. Collaborations between small companies or universities with materials or panel manufacturers can enable access to state-of-the-art materials and device optimization needed to demonstrate the longevity relevant to lighting applications. With the cumbersome work of device optimization in the hands of those with the materials, processes, and know-how to achieve best results, a material's potential can be satisfactorily assessed. There was resounding agreement amongst attendees that more partnership work would be beneficial and that complete materials solutions are essential.

2.3 Alternative Approaches

While there was agreement that there is a need for deeper understanding of materials properties and performance relationships as well as continued development work, some attendees promoted thinking outside the box to find solutions. Simultaneously stable and efficient blue emitters have been elusive for materials developers despite decades of work in the field. Though not proposing to abandon standard materials approaches, they suggested other possible ways of addressing the lifetime and efficiency shortcomings of blue emitters.

In particular, device architecture modifications can be used to enhance blue performance. For example, the device stack can be tailored to broaden the recombination zone which can lead to reduced excited state annihilation events. Molecular glasses could also be used to keep excited states apart and minimize roll-off. Another approach to longer lifetime is to simplify the device stack. By reducing the number of materials in a device or using materials that relax manufacturing requirements (e.g., neat films, single material hosts, low complexity emitter system, reduced number of transport/injection layers), more devices can be stacked in a tandem architecture without compromising manufacturability and cost. Such architectures allow much greater device lifetimes due to the lowered current density needed to achieve a desired luminance. One approach to stack simplification is to reduce the number of charge transport and injection layers, possibly through Fermi level engineering to enable ohmic contacts and improved charge generation layers for multijunction devices. Besides simplifying the stack, attendees suggested consideration of novel structures, such as quantum confinement for band-gap tuning and carrier confinement. Hybrid architectures were also proposed, such as using blue LEDs in conjunction with OLEDs to achieve the desired performance in diffuse light sources.

Novel thinking about ways to discover and approach materials was also proposed. For instance, using dilution molecules to alter properties of the organic semiconductors is a novel approach. This could be used to change the index of refraction of the OLED stack which could lead to significant light outcoupling improvements. For rapid materials discovery and design, the use of artificial intelligence (AI) tools and computational chemistry was proposed as a way of understanding the effects of aggregation.

Appendix A: Participant Presentations

Jian Li, Arizona State University: Blue Emitting Pt and Pd Complexes for Displays and Lighting Applications

Jian Li, Professor at Arizona State University, discussed the use of Pt and Pd complexes as replacements for Ir-based complexes in phosphorescent emitter systems. His team is investigating alternatives to Ir with the capability of increasing blue-green and amber emission with stable emitters. By designing molecules to shorten the emission lifetime, and thus reducing the opportunity for triplet-triplet annihilation and triplet-polaron annihilation, stability can be improved. The team has made significant progress with amber emitting Pd3O8-p materials, reducing lifetime from 200 to 0.62 µs while maintaining a photoluminescence quantum yield (PLQY) of close to 100%. These phosphorescent excimers have a planar structure which facilitates molecular alignment in the plane of the film translating to dipole moment orientation and improved light extraction efficiency. Best amber device performance was achieved with Pd3O8-py5 devices which showed an EQE of 36% at 1000 cd/m^2 (64% with optical matching glue to extract light trapped in substrate) with lifetime LT95 of 500 hrs at 18,000 cd/m^2. Blue emitting PdON3 structures are also being investigated. These molecules indicate a TADF-type emission and have a reduced emission lifetime of around 30 µs. Ultimately, the team is working to combine these Pd-based amber and blue emitters in a WOLED device structure. Preliminary results show the typical trade-off trend decreasing efficiency and stability with increased blue content, but all devices show remarkable color stability with current density. To conclude, Li suggested various topics for DOE R&D support including efficient and stable blue triplet emitters; cost-effective outcoupling enhancement; simpler white OLED structure (possibly OLED-LED hybrid devices); and low cost transparent anodes.

Peter Djurovich, University of Southern California

Peter Djurovich, Professor at the University of Southern California, presented on approaches to blue stability through molecular design. By reducing the emission lifetime, τ, device lifetime can be extended by the consequent reduction in triplet-triplet and triplet-polaron annihilation which lead to molecular degradation. After discussing the limitations of TADF and phosphorescence, he introduced his work on Cu, Ag, and Au based complexes which could be competitive alternatives to Iridium phosphors. Preliminary results from carbene-Cu-carbazolyl planar emitters showed that the emission can be easily tuned across the visible spectrum through the proper choice of acceptor (carbene) and donor (carbazolyl) groups. The compounds showed short decay lifetimes of ~1µs and very small energy difference between the singlet and triplet excited state (ΔE_{S1-T1}). Green OLEDs fabricated with these Cu complexes exhibited high EQE of ~20% and neat films were also efficient with EQE ~16%. Blue devices showed EQE as high as 12%. Djurovich went on to discuss work comparing Cu-based complexes to Ag-, and Au-centered complexes. Altering the metal from Cu to Ag or Au did not show any changes in emission color, but emission lifetime improvements were made. Using Ag which has the largest carbene – carbazolyl distance, τ was reduced to 0.33µs and 0.5µs for green and blue, respectively. Ag shows the fastest TADF rate primarily due to the very small energy difference between the singlet and triplet excited state. Djurovich proposed that further decrease in radiative lifetimes should be possible with reduced ΔE_{S1-T1}.

Tommie Royster, R-Display and Lighting: Advanced Blue Emitter Materials for OLED Lighting

Tommie Royster, CEO and Founder of R-Display and Lighting, emphasized the industry need for stable blue emitters. He presented his work on blue phosphorescent emitters for high efficiency lighting, leveraging molecular designs with high bond dissociation energy, with the ultimate goal of demonstrating high performance in a state-of-the-art OLED lighting stack. Recently, R-Display and Lighting discovered novel charge transfer emitters that can be modified to produce high efficiency phosphorescent emitters. By reducing symmetry of these materials, vacuum deposition properties can be improved. To conclude, Royster proposed research support from the DOE in the development of high triplet energy charge transporting and blocking materials (which are necessary for a stable blue stack) and funding of strategic partnerships to allow materials developers broader access to device fabrication and testing to screen for new materials.

Mike Molaire, Molecular Glasses: Molecular Distancing for Extremely Low OLED Roll-off Efficiency

Mike Molaire, CEO and Founder of Molecular Glasses, spoke on the issue of efficiency roll-off of WOLEDs at high brightness. Despite the dramatic growth of WOLEDs, low roll-off efficiency in high efficiency devices still needs to be addressed. He cited causes of efficiency roll-off, including non-radiative triplet exciton quenching, triplet-triplet annihilation, and triplet-polaron annihilation in the emissive layer. To suppress triplet exciton quenching, devices can be designed to balance charge carriers to minimize polaron accumulation at the interface. Further, decreasing the local triplet exciton density, extending the exciton formation zone, and shortening triplet exciton lifetime are strategies for improvement. Molaire then presented OLEDIQ™ materials whose high entropy of mixing helps to reduce triplet-triplet annihilation and concentration quenching, and emitter aggregation as they are non-crystallizable. When combined with charge carrier balance and an extended recombination zone, very low roll-off is expected. Indeed, when OLEDIQ hosts were compared to controls, an impressive reduction in efficiency roll-off was observed. For green emitters with EQE of ~18%, roll-off at 20,000 cd/m^2 was reduced from 12.2% to 7.9% in going from the control host to the OLEDIQ material. By using a mixed host for improved charge balance, a further reduction to 4.4% was achieved. At 60,000 cd/m^2, the roll-off efficiency was reduced from 27.0% (control) to 7.4% with a mixed OLEDIQ host.

Chris Giebink, Penn State University: Improving OLED Performance via Semiconductor Dilution

Chris Giebink, Professor at Pennsylvania State University, began by pointing out that some of the properties that are considered "intrinsic" to OLED materials could be modified for improved OLED device performance. Typically, OLED materials have a refractive index n ~ 1.7 which limits optical outcoupling efficiency. Further, glass transition temperatures are typically T_g ~ 100-120ºC, though OLED panels operate at elevated temperatures such that thermal stability is closely connected to catastrophic failure. Giebink then posed the question: "What if we could change properties like these without re-engineering OLED molecules?" He explained that by diluting the organic semiconductor with a carefully selected insulator, one has the opportunity to change optical and thermal properties without sacrificing electrical transport. He presented results of such an experiment using Teflon AF (n ~ 1.3) to dilute the hole-transporting N,N′-Di(1-naphthyl)-N,N′-diphenyl-(1,1′-biphenyl)-4,4′-diamine (NPD) organic semiconductor. For NPD hole-only devices, dilution lowered drive voltage and resulted in higher thermal stability. Further, it reduces built in voltage, improving carrier injection at the interface. Finally, bulk mobility decreases due to lower energetic disorder and percolation. Finally, Giebink presented the idea of tailoring the mobility and refractive index in every transport layer of a WOLED stack, with dilution molecules becoming a standard ingredient for OLEDs. Doing so may present an economic advantage by reducing the quantity of expensive stack materials required. Further, modeling shows internal extraction efficiency could be improved by 70% or more. However, a material other than Teflon is needed, and should be selected to optimize the electrical and optical performance as well as stability of devices.

Bernard Kippelen, Georgia Tech: Lighting in the 4th Industrial Revolution

Bernard Kippelen, Professor at Georgia Tech and Co-President of Institut Lafayette, considered what lighting in the future would look like, suggesting that the diffuse, large area, UV-free, ultra-thin, and transparent features of OLEDs make them an ideal choice for diffuse light sources. He proposed that lighting is part of an exciting technology revolution that culminates in the convergence of physical, biological, and digital impacts. Light can be used as more than a visible light source when combined with sensors, and that smart lighting presents a multi-functionality for value add. Further, there are biological impacts as demonstrated by circadian and melanopic lighting which influence metabolism and human health. Kippelen highlighted key R&D needs for OLEDs including smarter and greener materials (less usage of rare or noble metals), simplified device structures, manufacturing technology, and integration (packaging and bonding, including interconnects with backplane, barrier coatings, and functional films). Improvements in electrical contacts and charge generation layers for improved charge injection and electrical performance were proposed to help simplify device architecture and reduce the number of materials in the stack. Further investigation into novel structures, such as quantum-confined structures for carrier confinement and band-gap tuning, is also warranted. He then briefly spoke of his work on all-organic TADF compounds as an alternative to heavy metal containing phosphorescent molecules that are much more sensitive to concentration quenching.

Marina Kondakova, OLEDWorks: Material Development for OLED Lighting Panels Made by Vacuum Deposition

Marina Kondakova, Director and founding member of OLEDWorks, introduced the Brite 3 Family of OLED lighting panels. These are the brightest commercial OLED panels with up to 300 lm and 85 lm/W in warm (3000 K) and neutral (4000 K) white with CRI > 90 and R9 > 50. These panels meet industry's performance expectations in terms of output, efficacy, lifetime, and robustness, but they plan to step efficacy up to 100 lm/W and longer lifetime in their next generation of Brite offerings through the use of advanced materials for vacuum deposition and improvements in charge generation and electron injection layers, thermal stability, light extraction, and transparent conducting oxides (TCOs) with lower blue absorption. Kondakova summarized the strengths and weaknesses of trending blue emitter approaches including fluorescence, phosphorescence, TADF, and hyperfluorescence. She suggested that hyperfluorescent blue emissive layer (EML) seems the most promising approach for stable, efficient blue, but the components (host, emitter and/or TADF dopants) need to be developed in parallel to achieve the best performance. She noted there is room for improvement in green and red emitters as well. For instance, longer lifetime green-yellow phosphorescent emitters and narrower red phosphorescent emitters are needed along with host materials (preferably single material for best performance in manufacturing) that allow for improved efficiency and lifetime. Improvements in charge injection and transport layers could allow simultaneously a reduction in absorption, voltage, and plasmonic losses. Kondakova concluded by emphasizing that any new materials need to be cost-effective and need to be tested in a state-of-the-art OLED lighting stack so that performance improvements can be verified.

Michele Ricks, EMD Performance Materials Corporation

Michele Ricks, Business Development Manager of OLED Materials for EMD Performance Materials, discussed key R&D needs for OLED materials. She agreed with other attendees that high efficiency blue lighting is the highest priority. Though this has been a long-standing challenge, she said some encouraging results were showing up. While pure TADF approaches may not be the solution, there were interesting ideas in hyperfluorescence, for instance, from the HyperOLED project funded by the European Union (EU). Ricks pointed out that many key challenges are shared with the display industry who is aggressively working for solutions. Some areas of research where results can be leveraged from the display industry include low voltage electron transport layers and host materials. Funding for OLED lighting could go to R&D topics that are exclusive to lighting such as development of low-absorption charge injection layers or to developing fundamental materials understanding to help solve high efficiency blue. In alignment with other attendees, Ricks agreed that partnerships in materials developments were key and can alleviate the burden of device optimization for researchers.

Eric Margulies, Universal Display Corporation: Phosphorescent OLED Materials

Eric Margulies, Senior Scientist at Universal Display Corporation, presented on phosphorescent OLED materials, highlighting the spectral considerations for OLED lighting. Fine tuning of the emission spectra of OLED materials could result in improved performance and efficiency. For quality lighting, the black body curve is emulated, though there are constraints. For blue light, a deep blue emission is desired for high CRI, but to avoid blue light hazard, the onset of blue emission should be tuned to eliminate this risk. For longer wavelengths, while efficacy gains can be made with reducing deep red emission, this needs to be achieved without sacrificing CRI and R9. Maximizing the green output while maintaining high CRI can lead to high efficiency devices. After discussing spectral considerations for OLED lighting, Margulies pointed to light extraction technology as one of the biggest R&D needs for OLED lighting. He emphasized the importance of considering lifetime in development of light extraction materials, saying that improvements need to be made, but not at the expense of device lifetime.

Ruiqing (Ray) Ma, Nanosys: Cd-free QDEL for High Efficiency Diffuse Light

Ray Ma, Senior Director at Nanosys, presented on QDEL as a diffuse light source. Ma contended that QDEL is the perfect diffuse light source as it offers high efficiency, low voltage, spectral tuning, high brightness, solution processability, and is low-cost. He presented a summary of recent advances in Cd-free QDEL technology, showing EQE over 20% for red and green and ~ 13% for blue. The main challenge for QDEL is lifetime. In a case study comparing OLED with QDEL, Ma pointed out that QDEL performed as well or better than OLED in all categories except lifetime which needs over an order of magnitude improvement. Ma then proposed three R&D topics for DOE funding: 1) blue lifetime with high EQE; 2) white QDEL structures; and 3) low-cost manufacturing. Towards the first goal of efficiency and stability for blue QDELs, work to eliminate crystal defects, impurities, and quenching by charge transport layers is needed for improved blue efficiency while efforts into particle shapes that allow stable facets, electrochemically stable ligands, and uniformity of shell coverage play into the device lifetime. Once high-performance blue is achieved, white QDEL structures can be pursued. Various device structures can be considered, including tandem, mixed emissive layer, and down conversion devices. Each have advantages and disadvantages, but tandem devices are most capable of achieving high efficiency and long lifetime. The challenge, said Ma, is to develop a solution processed tandem QDEL at low-cost. Since quantum dots are solution processable, they are amenable to many low-cost manufacturing techniques which will need to be developed for QDEL lighting.

Semion Saikin, Kebotix: Solution-Processed OLEDs with Supramolecular Aggregates

Semion Saikin, Chief Science Officer at Kebotix, discussed how his team can achieve rapid discovery and creation of advanced materials by combining data and AI with robotics. They are applying this approach to target the problem of aggregation in emissive layers, focusing on host-free and blue materials. Typically, when dopants aggregate, concentration quenching of the EML is observed. In some materials, however, the aggregation aids emission and neat films of these materials can be successfully used as efficient EMLs as has been shown with some TADF type emitters. The team is currently creating a library of dyes that show aggregation-induced emission and that are compatible with solution processing. The next step is to demonstrate prototype devices that exploit supramolecular aggregation of dyes.

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