

Advances in the LED Materials and Architectures for Energy-Saving Solid-State Lighting Toward “Lighting Revolution”

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Abstract: In this paper, we review the recent developments (in years 2010–2011) of energy-saving solid-state lighting. The industry of white light-emitting diodes (LEDs) has made significant progress, and today, white LED market is increasing (mostly with increasing LED screen and LED TV sales). The so-called “lighting revolution” has not yet really happened on a wide scale because of the lighting efficiency at a given ownership cost. Nevertheless, the rapid development of the white LEDs is expected to soon trigger and expand the revolution.

Index Terms: Solid-state lighting, light-emitting diodes, III-nitride.

Over centuries, artificial lighting technology has made an incredible progress from candles and gas and kerosene lamps to today’s incandescent and fluorescent lighting. As a result, the overall operating cost of light has been reduced by 4.3 orders of magnitude since the 1700s [1], [2]. However, this technological progress has also inevitably increased the consumption of light as the cost of light has decreased. Today, the world uses about 0.72% of its Gross Domestic Product (GDP) on light, for example, in 2010, World GDP of 63.12 T\$ (current U.S. \$), which means about 455 B\$ for artificial lighting. Additionally, as the cost of light has decreased and GDP per capita has increased, the consumption of light per capita has also increased (almost linearly) over time. Over the last three centuries, the consumption of light per capita has increased by 5.4 orders of magnitude. Today, this has reached such a high level that an average person in a well-developed part of the world is effectively surrounded by tens of 100 W light bulbs at all times during his/her waking hours. Therefore, today, too much artificial light is consumed, which costs too much energy. According to a 2006 report by the International Energy Agency (IEA) and the Organization for Economic Co-operation and Development (OECD), lighting is responsible for ~19% of electricity consumption and ~6% of carbon emission. For our modern civilization today, energy-saving lighting is therefore increasingly essential. Efficient artificial lighting could save billions of dollars and reduce

greenhouse gases, while improving consumer's vision, provided that the lighting is also photometrically high quality.

Solid-state lighting (SSL) based on light-emitting diodes (LEDs) has attracted enormous interest [3], [4] and is of great importance to the development of electric lighting technologies, over a century after the early invention of incandescent bulbs (circa 1879) and over half a century after fluorescent lamps in the 1930s. The energy consumption used for lighting can be in principle reduced by 50% using LED lighting (if the targeted performance is met) and even more (if smart lighting enabled by LEDs is used). This corresponds to substantial annual carbon emission reduction of many hundreds of millions of tons per year. However, the so-called "lighting revolution" of LEDs after Thomas Edison is yet to be realized at wide scale. The lighting industry still has hurdles to clear before LED lighting penetrates the consumer market at the wide scale. Beyond the very real technical issues including device operating luminous efficacy (the efficiency droop and thermal droop) and light photometric properties, cost remains one of the main challenges for the LED lighting industry. Fortunately, recent rapid developments in LED materials and devices have brought it one step closer to the wide-scale commercialization and adaptation of general lighting.

Since the first bright blue and white LED chips were successfully demonstrated, InGaN LED has been making tremendous achievements in setting new records for luminous efficacy. In early 2010, Nichia has taken the white LED's luminous efficacy a step further, achieving values reported as high as 249 lm/W at 20 mA and 183 lm/W at 350 mA with $450 \times 450 \mu\text{m}^2$ blue LED die [5]. For comparison, a conventional incandescent light bulb of 60–100 W emits around 15 lm/W, and standard fluorescent lights emit up to 100 lm/W. In just one year, the LED's luminous efficacy was significantly enhanced. In March 2011, Osram Opto Semiconductors in Germany have set a new record efficacy for a warm-white LED that shows an efficacy of 142 lm/W reported at a correlated color temperature (*CCT*) of 2755 K and a color rendering index (*CRI*) of 81 at 350 mA/mm² [6]. Not much later, the US-headquartered LED maker Cree has set the new benchmark for the LED's luminous efficacy. The laboratory performance of the "neutral white" LED showed 231 lm/W at an operating current of 350 mA [7]. The achievement approaches the predicted maximum luminous efficacy of 260–300 lm/W [8].

In order to supply higher luminous flux, LEDs need much larger die size to handle large input power. However, a recurring problem is that the LED's luminous efficacy falls sharply under elevated current injection, and yet high current operation is necessary in SSL. This effect is known as the efficiency droop [9]. If the droop can be minimized, then the operating LED efficiency increases, which means either brighter LED is obtained, or less power is needed to run the device at the required illumination level. Though the root-cause of the droop is still debatable, various models have been adopted to explain the droop, such as junction heating [10], electron overflow [11], [12], reduced effective radiative recombination rate due to the elevated plasma temperature caused by carrier-carrier and carrier-photon collisions [12], and Auger recombination [13]. To date, several methods have been proposed to reduce the efficiency droop and enhance the optical output power of LEDs that are grown on *c*-plane sapphire substrates. For example, Schubert *et al.* proposed quaternary AlGaInN quantum barriers to suppress the polarization discontinuity between the adjacent well, and barrier demonstrates the excellency in reducing the efficiency droop [14]. Zhao *et al.* also suggested $\text{Al}_{0.83}\text{In}_{0.71}\text{N}/\text{GaN}/\text{Al}_{0.83}\text{In}_{0.71}\text{N}$ as the quantum barriers to enhance the electron injection efficiency [15]. Similarly, $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ as the electron blocking layer shows its advantage over AlGaIn in preventing electron overflow [16]. Recently, it has been shown that AlGaIn/GaN/AlGaIn as quantum barriers is effective not only in carrier confinement but in enhancing the quantum efficiency as well, while keeping the exact emission wavelength for green LEDs [17]. Moreover, AlGaIn, serving as the cap layer, has significantly reduced the strain energy and misfit dislocation density in the quantum wells [18], and this feature has been applied to the LED with AlGaIn/InGaIn superlattice as the quantum barriers and the electron blocking layer [19].

In addition to improving the carrier injection efficiency, it is also vital to increase the spatial overlap between the electron and hole wave functions. Staggered quantum well [20]–[23] and type-II quantum well architectures [24] were proposed for both LEDs and laser diodes for this purpose. The improved spatial overlap between electron and hole wave functions can also be achieved by

inserting a “ δ ” layer into the quantum wells. AlGaIn [25], GaN [26], and InN [27], [28] with an optimized thickness were employed to manifest the essentiality of deepening the localized states for better carrier confinement [29], [30]. In the meantime, the lattice mismatch between InGaIn and GaN can also be released in the quantum dots (QDs), which leads to a better electron–hole overlap and increased radiative recombination rates [31]. InGaIn/GaN wafers grown nonpolar/semipolar orientation have eliminated the polarization-induced fields [32]–[34], therefore enabling a complete overlap between electron and hole wave functions and an increase in the optical matrix element [35], [36].

Other than the record-breaking luminous efficacy levels, LED makers also devoted great effort to bring down the fabrication cost and, hence, the cost of light. GaN-based LED epitaxial on large-area Si substrates has been a hot topic in recent years. However, the intrinsic mismatch in both lattice constant and thermal coefficient of expansion with the requisite GaN epitaxial films challenges the epitaxial growth and hence the performance of the GaN-on-Si LEDs. The threading dislocations are expected to result in poor efficiency of the resulting LEDs while the macroscopic film stresses are to yield wafer bowing and even cracks. Nevertheless, significant progress has been reported in 2011. In March 2011, California-based LED maker Bridgelux Inc. has demonstrated a 135-lm/W LED on silicon substrate that was operated at 350 mA [37]. By using low-cost silicon substrates, instead of conventionally used sapphire or silicon carbide substrates, Bridgelux claimed it can deliver a 75% reduction in cost, which is one step closer to commercialization. More recently, Osram Opto Semiconductors has also announced that it is in the pilot stage of producing LEDs on silicon. The prototypes exhibited 127 lm/W at 350 mA according to the reports [38]. Other than GaN-on-Si, recently, Zhang *et al.* proposed numerically the growth of III-nitride LEDs on InGaIn ternary substrate that gives rise to higher spontaneous emission rate and reduced blue-shift of peak emission wavelength [39]. Besides, with proper indium composition, the lattice constant of InGaIn substrate is matched with that of multiple-quantum-well to be grown on top of it, leading to a reduced threshold carrier density and, hence, suppressed Auger recombination [40]. This provides a possibility of applying the InGaIn substrate for the development of efficient blue, green, and red high-brightness LEDs and laser diodes.

On the other hand, many LED makers started to emphasize the vertical LED architectures due to their many advantages over transverse or lateral counterparts [41]–[44]. The vertical LED architectures, involving removal of nonconductive (electrical and/or thermal) substrates, allow for better homogenous current distribution through the active region. Moreover, with the wider choice of receptor wafers (mostly metallic thin films), heat can be well dissipated and managed and, thus, can facilitate high-power LED operation. In the case of GaN LEDs on sapphire substrates, a nondestructive excimer laser-assisted lift-off of the sapphire substrates as a whole could open up the window for recycling of the sapphire substrates for epitaxial growth. This is a significant cost-saving aspect in the LED fabrication. On the other hand, instead of fully removal of sapphire substrate via excimer laser-assisted lift-off, Yang *et al.* reported the technique of thinning the substrate and etching down to the n-GaN region for metal contact, which also leads to enhanced output power and reduced forward voltage compared with the conventional lateral LED on sapphire [45]. The vertical LED architectures enable high power operation, thus increasing the maximum total flux output that can be obtained from a single chip. The latter factor also relates to cost, as it reduces the number of LED chips to achieve the same level of illumination. The aggressive device architecture advances in the GaN LEDs is expected to have an extremely beneficial impact in driving down the cost for GaN-based LEDs.

Optimization of light extraction efficiency is always a challenge for maximizing light output from the active region. Photonic crystals are one of the most commonly used techniques to solve this problem. Embedded air-gap photonic crystals show good performance in enhancing the extraction of polarized light emission as well as the polarization ratio [46]. Through pattern design of the photonic crystals, the far-field emission directionality can be controlled [47]. Meanwhile, the addition of a microlenses array on top of the device is capable of increasing the light extraction efficiency by more than two times [48]–[49] because of the increase in effective photon escape cone and a reduced level of internal reflection resulted from the grading of refractive index variation between the

GaN/SiO₂/PS/air interfaces. The surface plasmon offers another practical approach to overcome the optical output limit. The resonant coupling between the light-emitting excitons from the MQW region and the nanometer scale thickness metal layers enhance the luminescence intensity up to a factor of 5 [50]–[52].

Today, the most commonly used SSL sources are based on phosphor integrated color conversion LEDs, i.e., integrating of yttrium aluminum garnet (YAG) phosphors on blue InGaN LEDs [53]. The Y₃Al₅O₁₂:Ce³⁺ phosphor emits broad yellow light when subjected to blue light. It is commonly used as a coating on blue InGaN LEDs, converting part of the blue light into yellow, which then collectively makes white. Such an arrangement, however, yields white light generation with *CCT*s of 4000–8000 K, corresponding to the neutral and cool-white intervals, and *CRI*s typically lower than 80 [4], [54]. The low *CRI*s is due to the missing of red component in the spectrum. Sulfide phosphors such as (Ca_{1-x}, Sr_x)S:Eu²⁺ [55] and Y₂O₂S:Eu³⁺ [56] have been used as red phosphors for white LEDs for a long time. Unfortunately, their chemical stability is not desirable. Recently, various new red phosphor systems with good stability have been reported, especially nitride-based phosphors, including CaAlSiN₃:Eu²⁺ [57], Sr₂Si₅N₈:Eu²⁺ [58], Ba₂AlSi₅N₉:Eu²⁺ [59], and β-SiAlON:Pr³⁺ [60], exhibiting great potential as efficient red phosphors. However, for high-quality and wide-scale use in indoor illumination applications, white LEDs are required to provide warm enough *CCT* (< 4000 K) with high enough *CRI* (> 80) [4], [54]. Although a red phosphor can be added (triple-color mixing), the phosphor based color conversion LED suffers from the intrinsic problem that the emission spectrum is very broad with an inevitable far red emission spilling beyond the eye sensitivity curve. This problem leads to a fundamental tradeoff between the luminous efficacy of optical radiation and *CRI*.

Over the past two decades, a series of experiments sponsored by the US Department of Energy showed that rod photoreceptors responsible for scotopic (dark adapted) vision, as traditionally understood, also play an important role in vision in typical lighting conditions (e.g., offices, homes) [61]. Rods are responsible for the monochromatic vision in low light, which is called scotopic vision, as opposed to the photopic (photon adapted) vision of color perception provided by cone eye cells that are nonfunctional in low light. According to these reports, the tests showed that *a*) the rod photoreceptors are active at typical interior light levels, *b*) human eye pupil (which affects the acuity of our vision) is primarily controlled by rods (scotopic response), and *c*) the brightness of a light source is determined by both the photopic and scotopic responses. Also, according to a test carried out by Intel Corporation in Oregon [62], changing from normal lighting to scotopic enhanced lighting resulted in a total energy reduction of 57% while maintaining the same perceived brightness. However, in current practice, light sources are typically designed for photopic vision. To account for the rod response, the scotopic/photopic ratio (*S/P* ratio) is required. The *S/P* ratio of common light sources generally ranges from 0.8 to 2.5 (e.g., incandescent light with 1.41 and sunlight with 2.47) [63]. The commercially available YAG phosphor white LEDs exhibit poor *S/P* ratios typically ranging from 1.68 to 2.38 [64]. For single sources (including the state-of-the-art single-chip LEDs), the *S/P* ratio is limited to < 2.5.

The problems of low *S/P* ratio lighting and low *CRI* can be solved by modifying the phosphors. This requires design and synthesis of new phosphors, for example, by using nanophosphors of colloidal semiconductor QDs [65]. Due to the size tunability and narrow emission bandwidth (~30 nm), QD luminophors in white LEDs enable precise spectrum engineering to realize high-quality SSL tailored for different lighting requirements. Moreover, colloidal QDs feature large photoluminescence (PL) quantum yield in solution (reasonable yield in film) and increasing absorption below its emission peak toward shorter wavelengths. In fact, warm-white LEDs integrated with colloidal QDs with desirably low *CCT* < 3000 K and high *CRI* close to 90 have been achieved [66], [67].

In summary, a great opportunity for the LED-based SSL is presented to significantly impact the sociological, economic, and environmental future. Though, at present, the market demand for LED is currently dominated in flat panel display industry, given the continuous development in high-brightness LEDs, the LED-based SSL is in the limelight.

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