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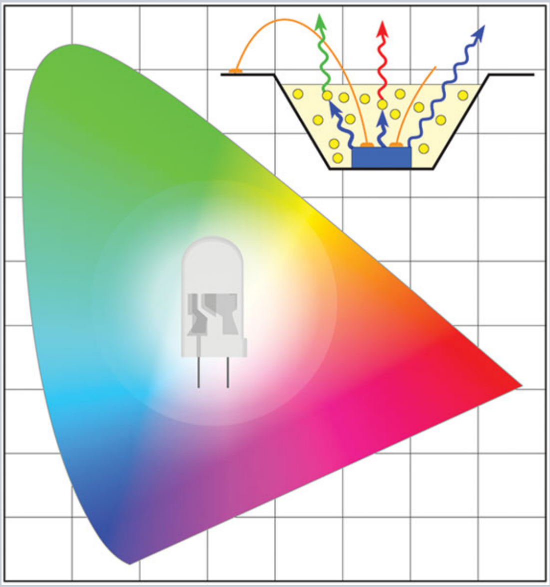
LASER & PHOTONICS REVIEWS

Abstract About twenty years ago, in the autumn of 1996, the first white light-emitting diodes (LEDs) were offered for sale. These then-new devices ushered in a new era in lighting by displacing lower-efficiency conventional light sources including Edison's venerable incandescent lamp as well as the Hg-discharge-based fluorescent lamp. We review the history of the conception, improvement, and commercialization of the white LED. Early models of white LEDs already exceeded the efficiency of low-wattage incandescent lamps, and extraordinary progress has been made during the last 20 years. The review also includes a discussion of advances in blue LED chips, device architecture, light extraction, and phosphors. Finally, we offer a brief outlook on opportunities provided by smart LED technology.



REVIEW ARTICLE

White light-emitting diodes: History, progress, and future

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1. History of white LEDs

Traditional light-emitting diodes (LEDs) emit monochromatic light. However, white LEDs, by definition, emit polychromatic light. Therefore, white LEDs are a significant departure from traditional LEDs. In the spirit of this significant departure, the phrase "solid-state lighting" is frequently employed for the field of white LEDs. While traditional LEDs, i.e. monochromatic LEDs, created mostly their own new markets, the implication of the phrase "solid-state lighting" is that LEDs are used to replace conventional lighting sources: Incandescent lamps (Thomas Edison's light bulb) and fluorescent lamps. Therefore, the phrase "solid-state lighting" is employed for white LEDs that are used in applications traditionally served by conventional white-light sources (incandescent and fluorescent lamps). Solid-state lighting includes general lighting in homes and offices, street lighting, automotive lighting, and backlighting in liquid-crystal displays (LCDs).

High-efficiency red, orange, and yellow devices (AlGaInP LEDs) had been developed in the 1980s. High-efficiency violet, blue, and green devices (GaInN LEDs) had been developed in the early 1990s. Therefore, in the mid-1990s, monochromatic high-efficiency devices, covering the entire visible spectrum, were available. As a result, the generation of white light by LEDs had been enabled. There are several viable approaches for white LEDs:

A ***first approach*** is a multi-LED-chip approach in which the light emitted from three LED chips emitting the three primary colors (red, green, and blue) is mixed to generate white light [1]. This approach is illustrated in Fig. 1. Inspection of the figure reveals that the lamp needs to have four lead electrodes so that each of the three LED chips can be injected with an appropriate current. The approach became feasible with the demonstration of highly efficient blue and green LEDs [2, 3]. Nakamura *et al.* [3] stated: "By combining high-power and high-brightness blue InGaN SQW [single quantum well] LED, green InGaN SQW LED and red GaAlAs LED, many kinds of applications, such as LED full-color displays and LED white lamps for use in place of light bulbs or fluorescent lamps, are now possible with characteristics of high reliability, high durability and low energy consumption."

A ***second approach*** is based on using a ultraviolet (UV) or violet LED chip and a phosphor that absorbs the UV or violet light and converts it to a broadband white light. Due to its similarity to conventional fluorescent lamps [4], this approach has been proposed multiple times prior to the advent of GaN-based blue LEDs [5]. Tabuchi [6] disclosed an LED structure, shown in Fig. 2 that includes an LED chip and a

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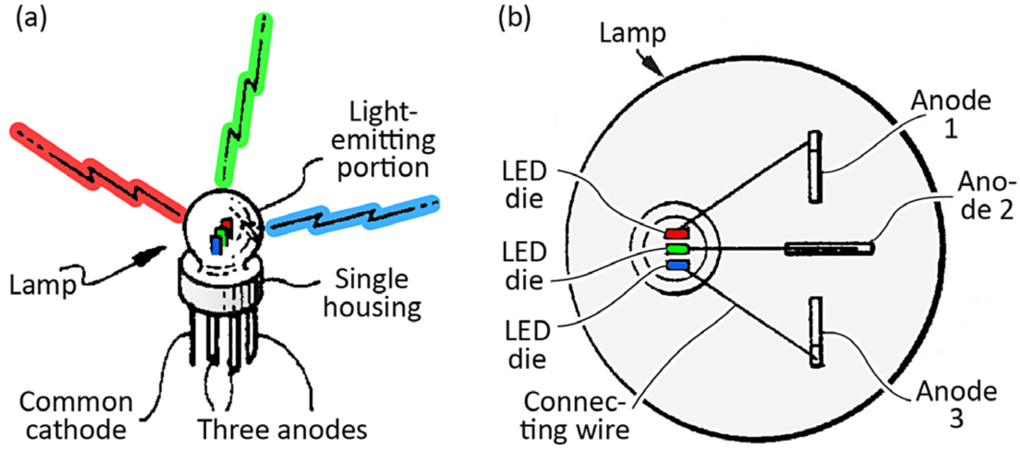


Figure 1 (a) Perspective view and (b) top view of multi-LED-chip white LED consisting of a red (R), green (G), and blue (B) LED chip (or die). Optical mixing of the RGB emission components results in white light (adapted from Ref. [1]).

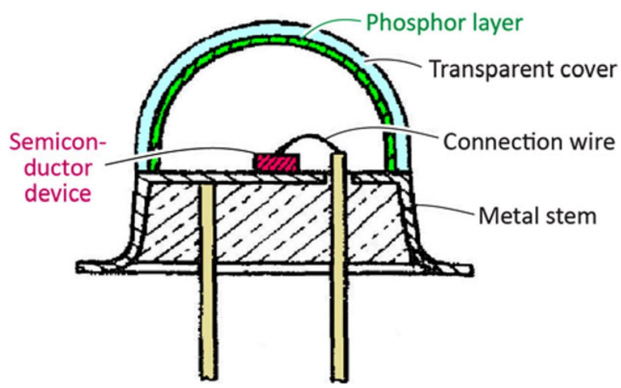


Figure 2 LED structure according to Tabuchi [6] having an LED chip exciting a phosphor that is coated on the inside of a transparent cover. The arrangement of the phosphor is reminiscent of a fluorescent lamp that has the inside of a glass tube coated with a phosphor (adapted from Ref. [6]).

phosphor that is coated on the inside of a transparent cover. Tabuchi [6] stated: "For example, it goes without saying that a near UV light emitting devices with GaN can be employed and that an ordinary UV-visible light conversion phosphor can be utilized." Another research group, Stevenson *et al.* [7] stated: "violet light [from an LED chip] may be converted to lower frequencies [. . .] using organic and inorganic phosphors. Such a conversion is appropriate [. . .] to produce light in a spectral range of greater sensitivity for the human eye. By use of different phosphors, all the primary colors may be developed [and] may be used for color display systems". Yet another research group, Tokailin and Hosokawa [8] disclosed using an organic UV LED and an organic phosphor converting the UV light to white light: "An electroluminescent element [. . .] which emits a near ultraviolet ray of light and a fluorescent material part which absorbs the ultraviolet light [. . .] and emits a fluorescence in a visible light range from blue to red [. . .] to be used as an element for emitting white light." That is, in each of these proposals, a current-injected pn-junction emitting near UV or violet light is used to excite a phosphor that emits visible light in the range from blue to red, i.e. white light. However, the proposals of Tabuchi, Stevenson *et al.*, and Tokailin and Hosokawa were not followed up by practical demonstrations. One may note that this approach can suffer from a low efficiency, because the down-conversion of UV or violet light involves a relatively large wavelength shift (large Stokes shift).

A ***third and most successful approach*** is a blue-LED-chip-plus-phosphor combination with the phosphor emitting green and red light. The blue light from the LED chip is *partially* absorbed by the phosphor while the other part of the blue light is transmitted through the phosphor. As a result, the blue light (from LED chip), green light (from phosphor), and red light (from phosphor) together form white light [9, 10]. This concept is now known as the "partial conversion" concept. Shimizu [9] stated: "A part of the [blue] light is absorbed by the phosphor, its wavelength is converted at the same time, and then the light is radiated." After an extensive search, numerous tests, and after the realization that organic phosphors could not operate reliably under the harsh operating conditions of a blue LED, a very suitable inorganic phosphor was identified by Shimizu *et al.* [11, 12]: Cerium-doped yttrium-aluminum garnet phosphor (YAG:Ce). This phosphor is able to absorb blue light and emits red and green light [13]. The approach, first demonstrated by Shimizu *et al.* [11, 12], was further described by Bando *et al.* [14, 15] and subsequently reviewed by Nakamura and Fasol [16]. The approach evolved into a pervasive commercial success, in part due to its efficiency, simplicity, and the requirement of only one LED chip driven by only one power supply.

For phosphor-based white LEDs, it is useful to distinguish between *full conversion* and *partial conversion*. As shown in Fig. 3(a), for full conversion, all primary radiation (exciting radiation from the LED chip) is converted to secondary light by means of the phosphor. If the primary radiation is UV light, the full conversion concept is

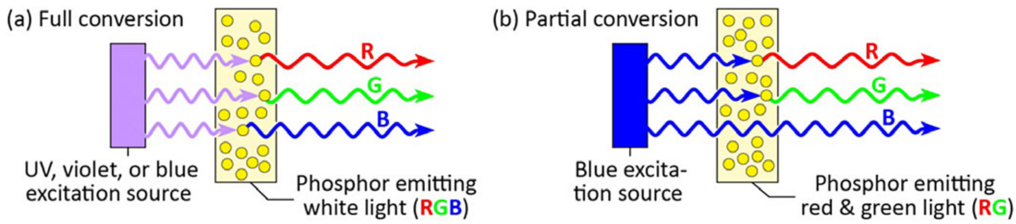


Figure 3 (a) Full conversion and (b) partial conversion of the excitation light in a white LED. In the case of full conversion, all primary light (excitation light) is converted to phosphor-based white fluorescence. In the case of partial conversion, the primary blue light becomes part of the white light.

reminiscent of the conventional fluorescent lamp in which all primary radiation (Hg-vapor radiation) is converted to secondary radiation. Tabuchi [6], Stevenson [7], and Baretz and Tischler [17] proposed to use the full-conversion concept to produce white light. In this case, the phosphor itself must emit white light. Figure 3(b) shows partial conversion in which only a part of the primary light (usually blue light) is converted to red and green light. In this case, the phosphor emission can be limited to red and green light, while the blue light is generated by the LED chip. Partial conversion has a fundamentally higher efficiency, since it inherently involves lower wavelength-conversion losses (lower Stokes-shift losses). Shimizu *et al.* [11, 12], Schlotter *et al.* [18], Hide *et al.* [19], and Reeh *et al.* [20] demonstrated the partial conversion concept for white LEDs.

One may note that the concept of combining an LED chip with a phosphor dates back to 1970 when Potter *et al.* [5] demonstrated the up-conversion of infrared (IR) radiation emitted by a GaAs LED chip to visible light by using a phosphor. Figure 4 shows (a) a GaAs LED chip mounted on a header and (b) the same chip covered by a blob of LaF_2:Tm or $(La_{0.8}Yb_{0.2})F_2$:Tm phosphor dispersed in a binder, polystyrene [5]. In the up-conversion process, the exciting photon energy is smaller than the phosphor-emission photon energy, that is $h\nu_1 < h\nu_2$, so that two exciting photons are required per phosphor-emitted photon. The up-conversion process is inefficient, even when used in conjunction with a high photon density. The down-conversion process ($h\nu_1 > h\nu_2$), used in white LEDs, is generally much more efficient.

During the year 1996, several publications and patent filings emerged that illustrate the pursuit for the white LED. Notable contributions to the field of white LEDs are, in chronological order:

- January 12, 1996: Shimizu [10] of the Nichia Company published a Japanese patent application publication titled "Sheet-like light source" that disclosed a planar white light source based on a blue LED and a red-emitting and a green-emitting phosphor for RGB white backlighting in full-color displays. Shimizu's white light source is the first demonstration of the partial-conversion concept.
- March 26, 1996: Baretz and Tischler [17] of the ATMI Company filed a patent application that discloses a semiconductor LED chip and a phosphor with the light emitted by the chip being fully converted by a phosphor so as to generate white light.
- June 26, 1996: Reeh et al. [20] of the Osram Company and the Fraunhofer Society, filed a patent application that discloses the partial conversion of blue LED-chip light by a yellow organic phosphor. However, as will be discussed below, the stability of organic phosphors is insufficient for LED applications.
- July 29, 1996: Shimizu and co-workers [11] at the Nichia Company filed a Japanese patent application that uses a garnet phosphor, specifically yttrium aluminum garnet, $Y_3Al_5O_{12}$, doped with the optically active rare-earth atom cerium (YAG:Ce). When excited in the blue wavelength range (e.g. at 450 nm), YAG:Ce has a broad emission band that spans from green to red. The YAG:Ce emission appears yellow to the observer, since the mixing of green and red light appears yellow to the human eye. Accordingly, YAG:Ce is frequently referred to as a yellow phosphor. Shimizu et al. [11, 12] demonstrated what would become a pervasively successful strategy in white LEDs: A blue LED-chip, the use of partial conversion, and the YAG:Ce phosphor. Some of the chip's blue light is converted to green and red light while the remaining part of the blue light is transmitted through the phosphor so that the blue, green, and red lights mix to form white light. The YAG:Ce phosphor exhibits superior chemical stability and can withstand the high radiation intensity of the blue LED chip. As the inventors pointed out, the YAG:Ce phosphor has to endure a blue light intensity that exceeds the intensity of sunlight by more than a factor of ten. The Sun's radiation intensity is about 1 kW/m^2; however, the radiation intensity of a blue LED can be 100 mW/mm^2 = 100 kW/m^2, i.e. 100 times higher than the radiation intensity of the Sun! YAG:Ce proved to be able to operate under such unprecedentedly harsh conditions. This is in contrast to organic phosphors that are generally unsuitable for operation under such harsh conditions [11,12]. Furthermore, Shimizu et al. [11,12] found that white LEDs using pure YAG:Ce has an unpleasant green-yellowish tint. Accordingly, the inventors worked to chemically modify the $Y_3Al_5O_{12}$:Ce by adding gadolinium, $(Y_{1-x}Gd_x)_3Al_5O_{12}$:Ce, as well as gallium, $Y_3(Al_{1-x}Ga_x)_5O_{12}$:Ce. Addition of Gd shifts the peak emission wavelength to longer wavelengths, thereby giving the phosphor a stronger red component. Similarly, addition of Ga shifts the peak emission

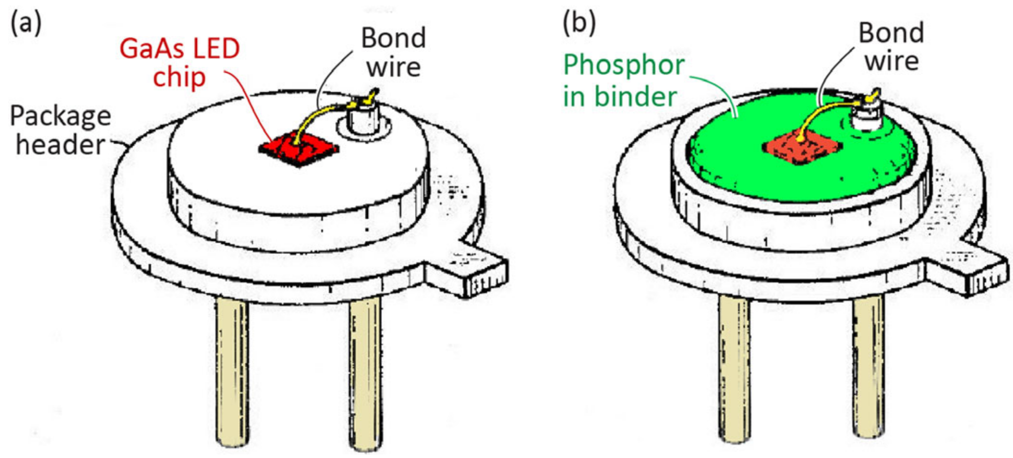
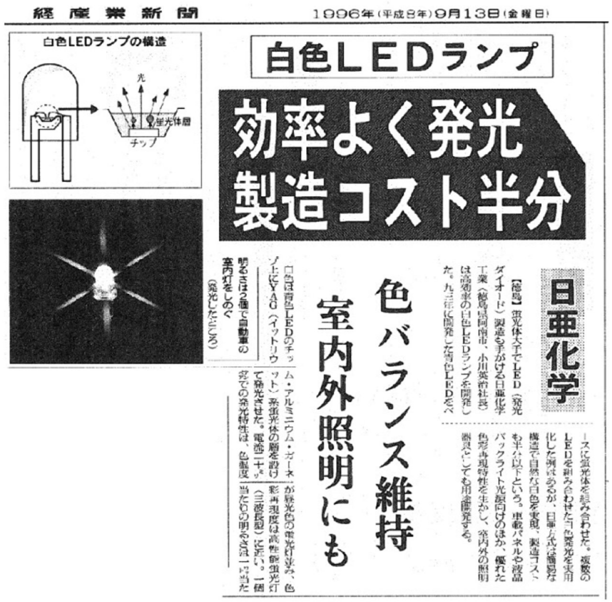


Figure 4 (a) GaAs-based infrared (IR) LED chip mounted on header. (b) LED chip coated by a phosphor dispersed in a binder for up-converting IR radiation to visible light (adapted from Ref. [5]).

wavelength to shorter wavelengths, thereby giving the phosphor a stronger green component. This allowed the new white LED to have a variety of color temperatures including daylight white (5 000 – 6 500 K) as well as incandescent-like white (2 500 – 3 300 K). Shimizu at al. [11, 12] can be considered the first disclosure of a viable white LED. The first white LEDs manufactured by the Nichia Company had a luminous efficacy of 5 lm/W; white LEDs (laboratory samples) with efficiencies as high as 12 lm/W were demonstrated by the team in 1996 [11]. This was a remarkable result: It was a world record for the efficacy of white LEDs and exceeded the efficiency of low-wattage incandescent lamps! The "blue-LED chip plus YAG:Ce phosphor" approach is still being practiced at the present time, decades after the invention.

- On September 13, 1996, a new era in lighting began: An article in the Japanese newspaper Nikkei Sangyo Shimbun [21], shown in Fig. 5, announced a new type of white light source, based on a blue LED and containing a yellow YAG:Ce phosphor. The white LED was reported to be efficient (5 lm/W), low cost, and have a predicted lifespan of 50 000 hours. What was announced in a relatively short newspaper article was a novel light source that was set to revolutionize the world of lighting.
- September 20, 1996: Reeh et al. [20] of the Osram Company filed a patent application that includes partial conversion of blue-LED light and the use of YAG:Ce phosphor.
- October 1996: Full-scale production of the phosphor-converted white LED started at the Nichia Company in Anan, Japan.
- November 29, 1996: Technical details of Nichia's white LED were presented at a technical meeting of the Institute of Phosphor Society (Japan) and the associated 264th Proceedings of the Institute of Phosphor Society (Japan). Nichia's phosphor-converted white LED, including its emission spectrum, is shown in Fig. 6 [14]. The emission spectrum is continuous covering nearly the entire visible wavelength range. Compared with the "spiky" emission


1996年(平成8年)9月13日(金曜日)

白色LEDランプの構造

白色LEDランプ

効率よく発光
製造コスト半分

日亜化学

色バランス維持
室内外照明にも

Figure 5 Article of the Japanese newspaper Nikkei Sangyo Shimbun on the impending commencement of manufacturing of white LEDs by Nichia Company: "White LED lamp: Light emission with high luminous efficiency halving production costs". The white LED is based on a blue LED chip and a YAG phosphor. A start of full-scale production of the white LED in early October 1996 is announced. A lifespan of 50, 000 hours or more is predicted, 10 and 50 times longer than the lifespan of fluorescent lamps and incandescent lamps, respectively. The white LED has a reported efficiency of 5 lm/W. The English translation of the article can be found in the Supporting information (from Ref. [21]).

spectrum of the then-common compact fluorescent lamps (CFLs), the quality of the white light emitted by the LED was superior. Likewise, compared with the power consumption of the then-common incandescent light bulbs, the prospect of white LEDs, in terms of power consumption, was far superior. Thus, the foundation of a new

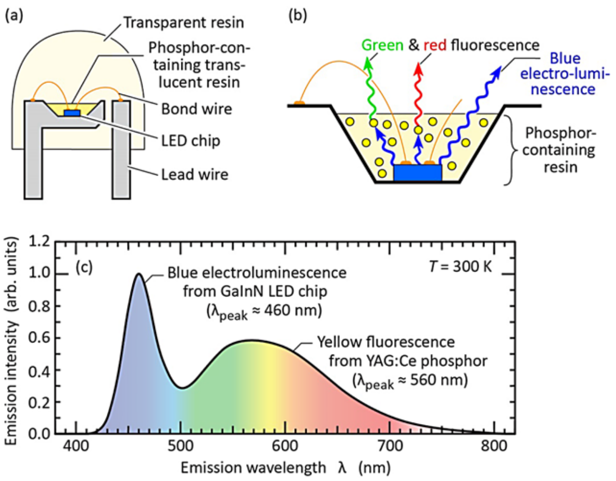


Figure 6 (a) Structure of the first white LED. (b) InGaN LED chip emitting blue electroluminescence (EL) and YAG:Ce phosphor, excited by blue EL, emitting broadband yellow fluorescence ranging from green to red thereby generating white light. (c) Emission spectrum of the first white LED showing distinct blue EL line at 460 nm and broad fluorescence band ranging from green to red (adapted from Ref. [14]).

LED white-light source, that was poised to replace conventional white-light sources, had been established.

- In the April 1997 issue of Applied Physics A, an Osram-Fraunhofer team [18] reported on blue-emitting GaN chips in combination with luminescence down-converting inorganic YAG:Ce phosphor as well as organic fluorescent dye. Using the LED-chip-phosphor combination, the team demonstrated LEDs emitting white light as well as mixed colors, such as cyan and magenta. Note that the similar concept was reported around the same time in the May 1997, in which the team used an InGaN/conjugated polymer hybrid LED for white emission [19].

Undoubtedly, a new era in the generation of white light started in the summer of 1996, based on semiconductor pn-junction diodes and phosphors. This innovation in the generation of white light can certainly be compared to the development of Edison's incandescent light bulb and the fluorescent tube that occurred decades earlier. Photographs of key innovators in the field of white LEDs are shown in Fig. 7, including Yoshinori Shimizu, Yasunobu Noguchi, and Kensho Sakano of the Nichia Company (Anan-Shi, Tokushima, Japan) and Peter Schlotter of the Fraunhofer Institute for Applied Solid-State Physics (Freiburg, Germany) [18].

In retrospect, it is easy to forget that there was substantial skepticism regarding white LEDs. There were numerous concerns about white LEDs, particularly from the established lighting community. Opinions such as "Poor color rendering of objects, narrow output cones, and lower total lumen outputs make solid-state lamps impractical for general illumination" were common in the early years of white LEDs [23]. The view that LEDs were to be relegated to niche applications was not uncommon either: "The limitations of commercial LEDs do not preclude their use in niche applications" [23]. Even in 2008, when it was abundantly clear that white LEDs are revolutionizing general lighting [24], strong skepticism was voiced in the New York Times: "I do not see a major step toward change in general illumination. To say LEDs will change everything, I don't [believe] it. I think a lot of it is hype" [25].

2. Progress in the efficiency of white LEDs

Traditional LEDs were used for indicator lights, signage, and displays. Prior to about 2000, illumination, i.e. the general lighting of homes, offices, and streets, was not the major purpose of LEDs. This was about to change in 2000 when the vision that LEDs can be used for lighting applications, was discussed in detail for the first time.

A report issued by Sandia National Laboratories and authored by Haitz *et al*. [26], addressed, in a quantitative way, the potential energy savings enabled by LED lighting (called "semiconductor lighting" in the report). The authors concluded that the large-scale introduction of solid-state lighting enables that the decrease of the electrical power consumed by lighting by more than 50%, amounting to global annual electrical energy savings that exceed 1 000 TWhr. The authors also discussed the emission of the global warming gas CO_2 which is generated through the production of electricity from coal-, oil-, and natural gas-burning power plants. The authors concluded that the large-scale introduction of solid-state lighting and the associated electricity savings would reduce CO_2 emissions by annually approximately 200 Mt (Megatons). Additional analyses, that are consistent with the major conclusions of Haitz *et al.* [26], include discussions by Bergh *et al.* [27] and Schubert *et al.* [28].

For general lighting applications, the *total power* emitted by an LED is of importance. Whereas conventional light sources such as incandescent light bulbs can be easily scaled up to provide a high luminous flux, LEDs, historically (i.e. during the pre-2000 time) have been low-power emitters. The historical development of the luminous flux per LED package, measured in lumens, is shown in Fig. 8 [29–35]. The figure shows that the luminous flux per LED package had increased by about four orders of magnitude over a period of 1968–2000 and the trend has continued so far. For comparison, the figure also shows a 60-W-incandescent-bulb's luminous flux and approximate purchase price. The figure illustrates continued progress in the performance and manufacturing cost of LEDs. Note that the cost shown in the figure is just the purchase price of the lamp and does not include the cost for the electrical energy consumed by the lamp. The cost of the electrical energy of incandescent lamps typically is much higher than their purchase price. That is, efficient LED light sources have a significant energy cost advantage over incandescent lamps.

The advancement of LED efficiency has been compared to the advancement made in Si integrated circuits where the performance increase versus time has been characterized by

"Moore's law". This "law" states that the performance of Si integrated circuits doubles approximately every 18 months. In this context, the term "*Haitz's observation*", also called "Haitz's law", may be mentioned [26, 36–38]. Haitz found that the amount of light generated by one LED lamp package (luminous flux per lamp package, measured in lumen per lamp package) increases by a factor of 30 per decade [26, 29] or by a factor of 20 per decade [38], while the luminous flux generated by one LED lamp costing a certain price (measured in lumen/US$) increases by a factor of 10 per decade [38]. A graphical representation of these observations is shown in Fig. 9.

It should be noted that the data presented in Haitz's observation were gathered selectively and restricted to certain LED material systems, emission colors, color temperatures, and time frames. Haitz and Tsao [38] admitted that their selection of data points "is not very scientific". Indeed, the selectivity by which the data was gathered led to the visually convincing dependence shown in Fig. 9. Despite its shortcomings, Haitz's observation is a useful articulation of the long-term trend towards LEDs with higher luminous flux as well as the trend towards higher luminous-flux available for a given LED purchase price. Inspection of the figure reveals that for midrange-power surface-mount device (SMD) white LEDs (typically 0.5 W, but ranging from 0.25 W to 1.0 W), the typical performance-to-price ratio as expressed in lumen per US$ (lm/$) ranges from 500 to 1500 lm/$. That is, a midrange power SMD white LEDs emitting 100 lm currently (2016) cost about 10 US cents. For a retrofit LED light bulb, emitting 500 lm (equivalent to a 40 W incandescent light bulb), the cost of the associated LEDs is about 0.50 $.

Jeffrey Y. Tsao, a pioneer in solid-state lighting working at the US Sandia National Laboratories (Albuquerque, New Mexico), is a co-author of the Haitz *et al.* report [26]. Tsao pointed out the correlation between the cost of light, the consumption of light, and productivity of human society. He stated: "throughout its history, lighting technology has made tremendous progress: the efficiency with which power is converted into usable light has increased 2.8 orders of magnitude [i.e. by a factor of 631] over three centuries. This progress has, in turn, fueled large increases in the

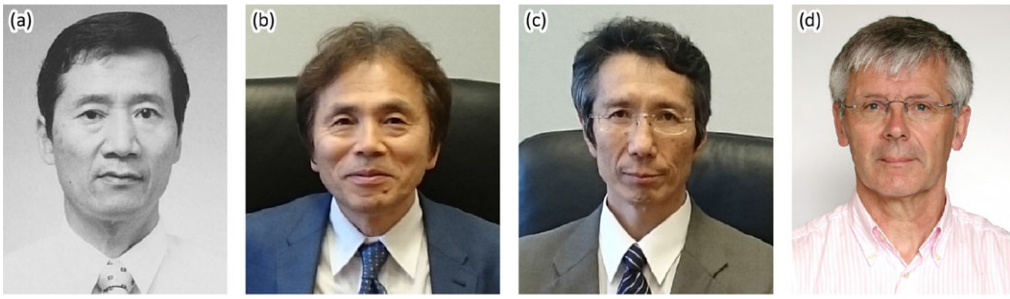


Figure 7 (a) Yoshinori Shimizu, pioneer of white LEDs and senior manager in the Nichia Engineering Department at Nichia [22]. (b) Yasunobu Noguchi, a pioneer who developed YAG:Ce phosphors for white LEDs. In 2015, he was head of a phosphor department at Nichia. (c) Kensho Sakano fabricated and tested hundreds of prototypes of the white LEDs. (d) Peter Schlotter (Fraunhofer Institute), first author of a highly-cited article on white LEDs [18] and co-inventor of a family of patents relating to white LEDs.

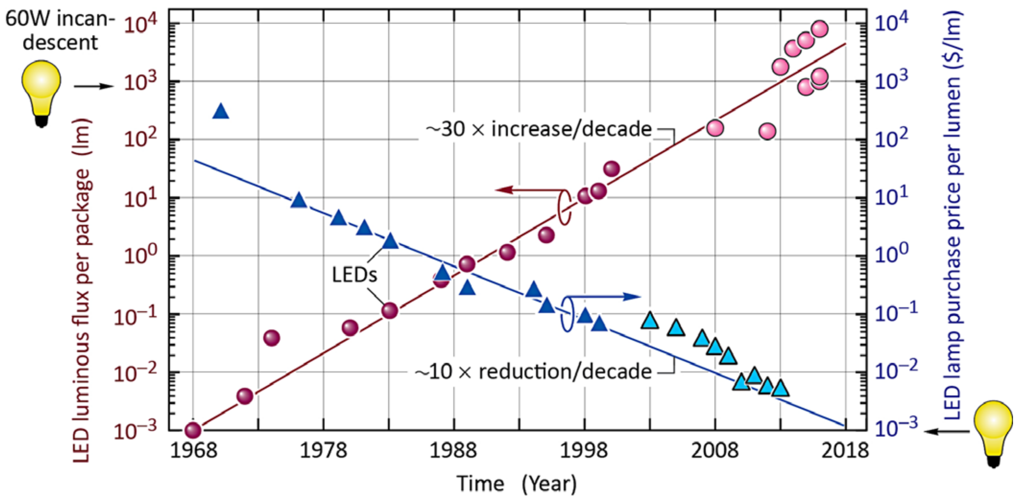


Figure 8 LED luminous flux per package and LED lamp purchase price per lumen versus year. Also shown are the values for a 60 W incandescent tungsten-filament light bulb with a luminous efficiency of ~17 lm/W and a luminous flux of 1000 lm with an approximate price of 1.00 US$ (adapted and updated from Refs. [29–35]).

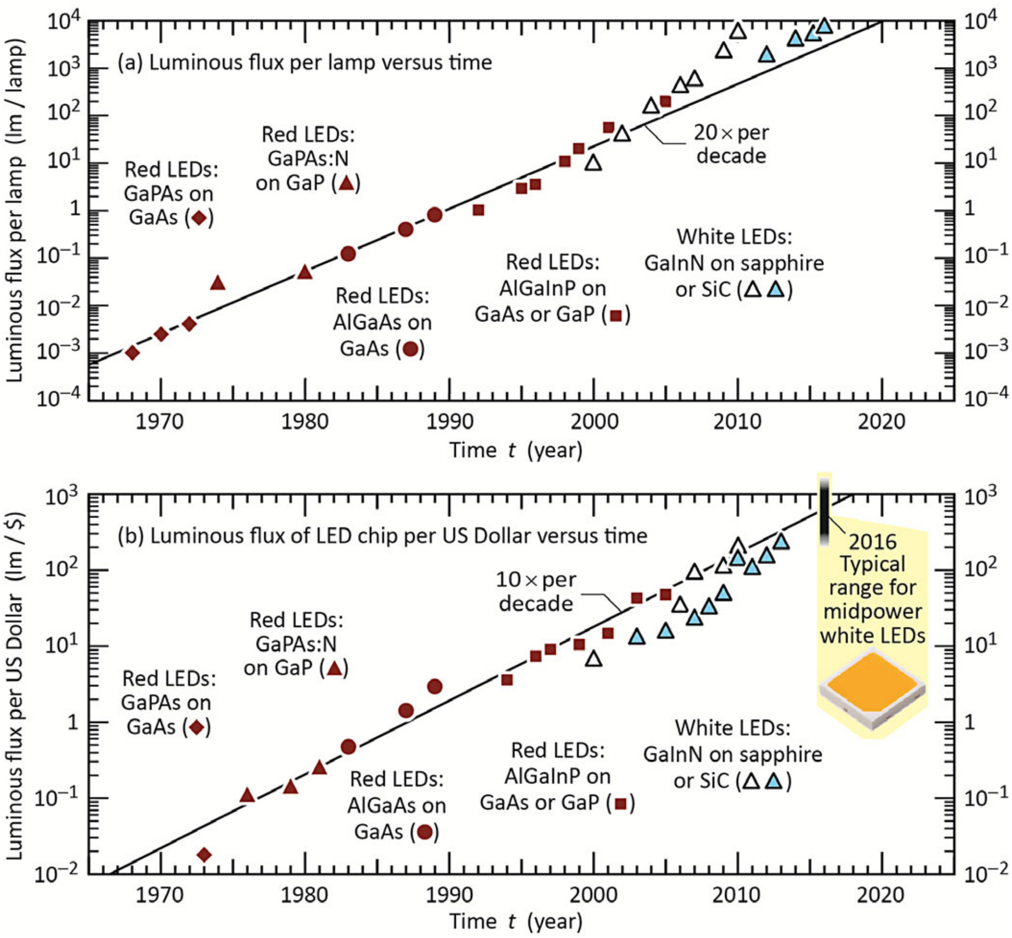


Figure 9 Development of (a) luminous flux per LED lamp and (b) luminous flux of LED lamp per unit cost (1.0 US Dollar) versus time (adapted and updated from Refs. [30–34], 38]]).

consumption of light and productivity of human society" [39,40].

The temporal evolution of the luminous efficiency of light sources is shown in Fig. 10. The figure shows the following light sources: Incandescent lamp with C (carbon) filament demonstrated in 1879 by Thomas A. Edison; Incandescent lamp with ductile W (tungsten) filament demonstrated in 1911 by William D. Coolidge; Linear fluorescent lamp introduced in 1937 at the New York World's Fair; CFL introduced in 1985 by Osram Company [41]. These now-obsolete light sources and their typical efficiencies are as follows:

- Incandescent light bulb with C (carbon) filament: 1.2 – 2 lm/W
- Incandescent light bulb with W (tungsten) filament: 10 – 18 lm/W
- Incandescent halogen light bulb with W (tungsten) filament: 16 – 24 lm/W
- Linear fluorescent lamp (LFL): 65 – 95 lm/W
- Compact fluorescent lamp (CFL): 50 – 70 lm/W

Subsequent to the invention and commercialization of the first white LED, the LED industry engaged in a race towards increasingly higher efficiencies. As mentioned previously, the first commercial white LED product, available in late 1996, had an average luminous efficacy of 5 lm/W with the best devices reaching 12 lm/W [12]. Subsequently, the value of 100 lm/W, which in the pre-2000 years was considered an astronomically high efficacy value for white LEDs, became a goal, a desired milestone, that could be called the "sound barrier" in the race for high efficiency in white LEDs. Conventional light sources that produce high-quality white light, e.g. fluorescent lamps, typically have efficacies below 100 lm/W. Demonstrating that LEDs can attain a greater-than 100 lm/W efficacy would mean that the days of the conventional white light sources are numbered and that their eventual demise would be inevitable. Would white LEDs be able to break the "sound barrier" of 100 lm/W? The barrier was indeed broken in 2004 by the Nichia Company, as reported by Narukawa [44]: "At a low current region (less than 1 mA), the luminous efficacy is estimated to be more than 100 lm/W, which indicates that InGaN-based white LEDs have great potential to replace fluorescent lamps (75 lm/W) in the near future." A couple

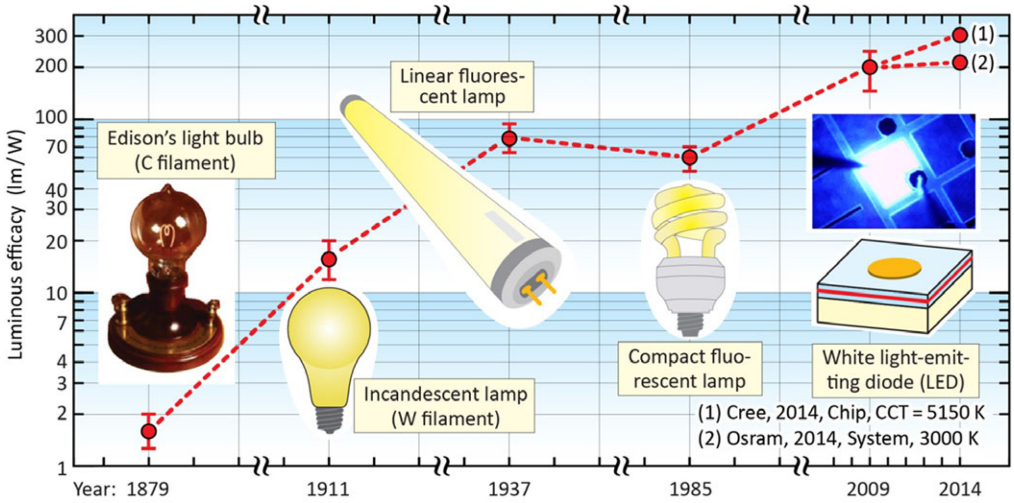


Figure 10 Temporal development of the luminous efficacy of different types of lamps. The 2014 points represent: (1) White LED device performance (Cree Company; 303 lm/W; CCT = 5150 K) [42], and (2) White LED device and system performance (Osram Company; 215 lm/W (device); 205 lm/W (system); CCT = 3000 K) [43].

of years later, in 2006, Narukawa *et al.* [24] reported efficacies as high as 174 lm/W at a low current of 2 mA (current density 1.98 A/cm^2) and 138 lm/W at a current of 20 mA (current density 19.8 A/cm^2, i.e. far in the droop regime). The "sound barrier" was not only broken, it was shattered by a wide margin making it abundantly clear that the demise of conventional light sources by new LED-based white light sources was inevitable and had begun.

In 2010, reported luminous efficacies of white LEDs were in the 150–250 lm/W range. Very high efficiencies were reported by several companies including the following: Nichia Company's Takashi Mukai announced a laboratory-result efficacy of 249 lm/W for a white LED injected with a very low current [45]. The Cree Company announced a laboratory-result efficacy of 208 lm/W for a white LED with a correlated color temperature of about 4 600 K at an injection current of 350 mA [46]. On March 26, 2014, the Cree Company announced a laboratory-result efficacy of 303 lm/W for a white LED lamp (excluding power supply) with a correlated color temperature of 5 150 K at an injection current of 350 mA [42]. On March 28, 2014, the Osram Company announced a lamp efficacy of 215 lm/W and a system efficacy (including power supply) of 205 lm/W for a white LED lamp system with a color temperature of 3 000 K [43].

The improvement in luminous efficiency of visible-spectrum LEDs has been truly breathtaking. The historical development of the luminous efficiency of visible-spectrum LEDs is shown in Fig. 11 [47–57]. The chart illustrates the modest beginnings of visible-spectrum LED technology which started in the 1960s. If the progress from 1960 to 2000 is assumed to be continuous and constant, then the LED luminous efficiency has doubled every 4 years. Figure 11 also shows the luminous efficiency of conventional (pre-LED) sources including Edison's first light bulb (1.4 lm/W) and red and yellow filtered incandescent lamps. The figure reveals that LEDs outperform filtered red and yellow incandescent lights by a large margin.

3. Technical features enabling progress in white LEDs

From the technical point of view, the improvement in the luminous efficiency of white LEDs has resulted from a series of breakthroughs and advances in crystal growth, heterostructures, device architecture, phosphors, and light outcoupling from GaInN-based *blue* LED chips. The 2014 Nobel Prize in Physics was awarded to Akasaki, Amano, and Nakamura for the invention and development of efficient GaInN blue LEDs. We now quote the citation of the 2014 Nobel Prize in Physics: "for the invention of efficient blue light-emitting diodes which has enabled bright and energy-saving white light sources". That is, the Nobel Prize was awarded for "the invention of efficient blue light-emitting diodes" and this invention has "enabled [LED-based] bright and energy-saving white light sources". The Nobel Committee recognized the interconnectedness of blue and white LEDs: "White LEDs used for lighting are often based on efficient blue LEDs that excite a phosphor so that the blue light is converted to white light. These high-quality LEDs with their very long lifetime (100 000 hours) are getting cheaper, and the market is currently exploding".

Firstly, lots of efforts were made to produce GaN single crystals by a hydride vapor phase epitaxy technique from the end of the 1960s. In a 1973 review article, Pankove identified two grand challenges [58]: "In spite of much progress in the study of GaN over the last two years, much remains to be done. The major goals in the technology of GaN should be: (1) the synthesis of strain-free single crystals, (2) the incorporation of a shallow acceptor in high concentrations." A breakthrough in the epitaxial growth of

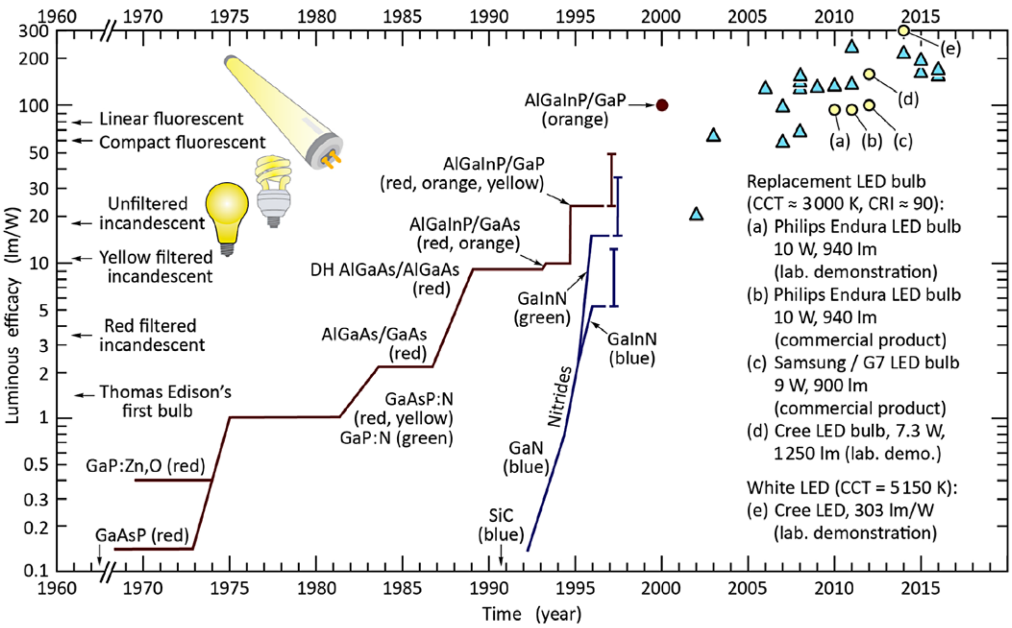


Figure 11 Development of luminous efficacy of visible-spectrum LEDs and luminous efficacy of conventional light sources (incandescent and fluorescent sources). The shown luminous efficacy values of LED retrofit light bulbs are from laboratory demonstrations and from commercial products (2002–2016) (adapted and updated from Refs. [47–57].

GaN with high crystal quality and good optical properties was indeed made by forming, at low temperature, a thin polycrystalline AlN nucleation (or buffer) layer on a sapphire substrate, followed by the epitaxial growth of GaN at high temperature using metal-organic vapor phase epitaxy [59]. The epitaxial GaN layer showed a highly smooth surface which was an important prerequisite for growing the thin hetero-structures that are needed for advanced LED structures. Various modifications of the buffer-layer growth technique followed to further decrease strain and dislocation density in the overgrown films [60, 61]. The difficulty of effective p-type doping in GaN has been another major issue for producing efficient p-n junction light emitters. Amano *et al.* discovered that the irradiation of Mg-doped GaN with a low energy electron beam results in p-type conductivity, a breakthrough that enabled the realization of p-n junction based blue LEDs [62]. Nakamura *et al.* showed that thermal annealing in a hydrogen-free atmosphere is an efficient method to de-passivate Mg acceptors [63, 64]. GaInN/GaN single- and multi-quantum well active regions were another notable accomplishment in the performance of blue and green LEDs [65–68].

A remarkable milestone in a GaInN blue LED was the finding that the efficiency of LEDs decreases with increasing operating current, a phenomenon called the "efficiency droop" [69–72]. This phenomenon is thought to originate from fundamental material properties of a GaInN material system. There are competing explanations for the efficiency droop in GaInN LEDs. There are a number of papers that support Auger recombination being the predominant cause of the droop [73, 74]. Other papers have identified the lack of hole injection as the predominant cause of the droop [72, 75, 76]. Lack of hole injection, is equivalent to electron leakage since for each hole not injected into the active region, there necessarily is one electron leaking out of the active region. The efficiency droop phenomenon is relatively complex so that a comprehensive discussion of the phenomenon goes beyond the scope of the present paper.

Secondly, various iterations in the device architecture combined with advanced process technology have diversified and improved the optical and electrical characteristics of LEDs. Most commercial LEDs are grown on electrically inert sapphire substrates, leading to the main device architectures shown in Fig. 12. Small-area (about 300× 300 μm^2), low-power LEDs typically adopt the conventional "epi-up" configuration, shown in Fig. 12(a). However, the epi-up configuration suffers from the inherent trade-off between good current spreading in the semi-transparent ohmic contact to p-type GaN and good light-extraction efficiency which is particularly compromised in high-power applications. That is, the higher the operating current is, the thicker must be the semi-transparent p-contact layer as required for good current spreading, which, however, increases the optical absorption loss in the semi-transparent contact layer. This problem can be alleviated by using the "flip-chip" architecture shown in Fig. 12(b) [36, 77]. The flip-chip configuration has the advantage of overcoming the above-mentioned trade-off by using a thick reflective metal p-contact so that better light-extraction efficiency, excellent dissipation of the heat generated in the active region, and a wirebond-free architecture is attained. Figure 12(c) shows an LED with vertical thin-film configuration employing sapphire substrate removal by an excimer laser, called laser-liftoff (LLO). This

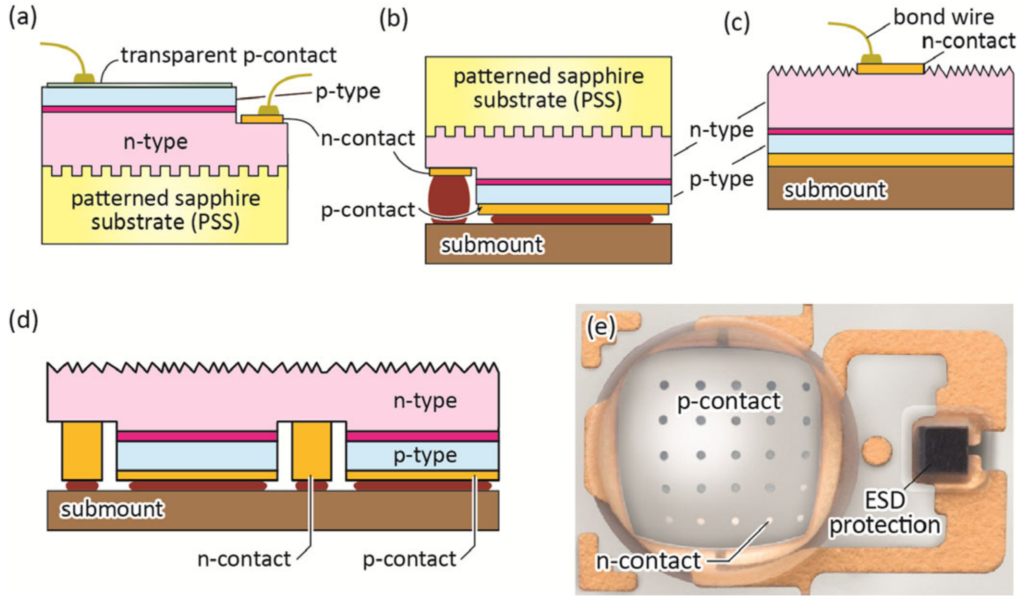


Figure 12 Typical chip architectures of GaN-based LEDs grown on sapphire substrates; (a) “epi-up” LED, (b) “flip-chip” LED, (c) vertical thin-film LED, and (d) buried p- and n-type contact structure. (e) Commercial GaInN blue LED employing buried contacts (after Ref. [81]).

configuration allows for the roughening of the n-type GaN N-face surface by means of crystallographic etching [78], a larger top-surface emitting area, and a thick reflective p-contact, thereby significantly enhancing light extraction compared to the epi-up and flip-chip architecture. However, the need for a wirebond to the n-contact complicates phosphor deposition. Moreover the usage of high-power excimer laser can lower the device yield of this architecture [79]. A design that combines the benefits of vertical thin-film and flip-chip configurations is the thin-film flip-chip (TFFC) architecture in which the sapphire substrate is removed by LLO after a flip-chip bonding process [80]. A time evolution in the extraction efficiency of LEDs with various device architectures was summarized by Krames *et al.* [79], in which the extraction efficiency of the TFFC LED was estimated to be 80%.

A further advancement over the TFFC configuration is the employment of an array of “buried” n-contacts as shown in Fig. 12(d). Inspection of the figure shows that both p-type and n-type contacts are located on the bottom side of the chip, i.e. they are buried under the chip. The voids between the contacts can be filled in by an electrically insulating underfill passivation layer (not shown in the figure). The p-type contact is optically reflective and light emission occurs through the roughened top surface. The elimination of the wirebond and the proximity of the active region to the chip surface allow for (i) phosphor deposition that does not interfere with any contact, (ii) close placement of secondary optics, and (iii) very effective heat dissipation. Figure 12(e) shows an example of a commercially available LED with buried contacts [81]. We note that buried contact LED configurations enable back-end processes (especially phosphor deposition) to be executed in a parallelized manner (rather than progressed in a serial manner, as is done in conventional packaging), i.e. full wafers or arrays of chips are processed together (in parallel) representing a significant cost advantage. The emerging field of chip-scale packaging (CSP) is based on such parallelization of LED fabrication processes [82].

Recently, researchers expanded the device architecture to flexible and stretchable applications [83–88]. For this purpose, epi-layer transfer methods have been developed, in which the transfer of a GaInN LED structure grown on a conventional substrate onto flexible or stretchable substrates is enabled by the help of a sacrificial layer. The most remarkable sacrificial layer reported had a form of quasi-2D layered structure, i.e. graphene, as shown in Fig. 13 [84]. Weak binding or van der Waals forces between adjacent layers in multilayered graphene allowed easy mechanical release of the overgrown LED thin-films and transfer to the target substrate, even to a plastic plate. Although the efficiency of the transferred LED presently is much lower than the conventional counterpart, this approach is believed to provide a new route for realizing flexible and stretchable blue, green, and white LEDs having high performance, low cost, and scalability.

Thirdly, one of the fundamental problems facing high-efficiency LEDs is the occurrence of trapped light inside high-refractive-index semiconductor materials. Total internal reflection at a flat interface between the semiconductor and the surrounding medium means only light emitted into a light-escape cone can escape from the LED chip [89]. Trapped light will eventually be reabsorbed, for example, by the substrate, active region or metal contacts, reducing the efficiency of an LED. In order to extract more light from an LED chip, various efforts have

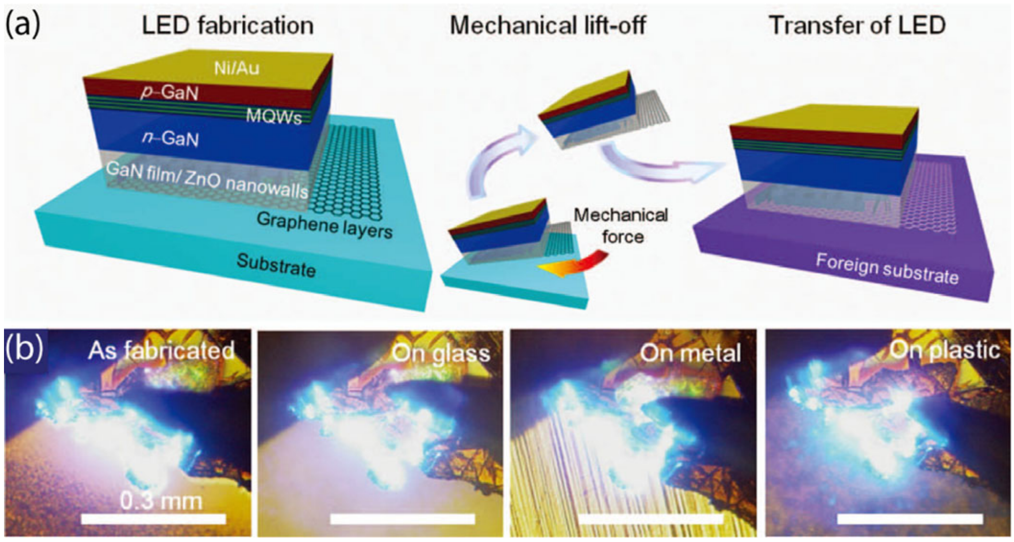


Figure 13 (a) Schematic illustration of the transfer processes for thin-film LEDs grown on graphene-layer substrates. (b) Optical images of light emissions from the as-fabricated LED on the original substrate, and transferred LEDs on the foreign metal, glass, and plastic substrates. (after Ref. [84]).

been made such as shaping of LED dies [90], texturing of semiconductor surfaces and interfaces [91], and using highly reflective mirrors and anti-reflection optical coatings [92]. Die shaping such as pedestal-shaped LEDs [93], truncated-inverted pyramid LEDs [94], truncated-pyramid-bonded LEDs [95], and recently triangular-shaped LEDs [96] have been demonstrated to enhance light extraction over the conventional rectangular parallelepiped LED dies. However, die shaping technologies are becoming less interesting because of the trend towards thin-film LEDs. Accordingly, the main method to enhance light extraction is, as discussed above, the roughening or texturing of semiconductor surfaces by means of crystallographic etching. This process allows for efficient out-coupling of the light by scattering at the chip surface. Additional ways to reduce optical losses in LEDs include the employment of reflectors and optical coatings with anti-reflection characteristics on the top surface [97,98]. Kim *et al.* [98] demonstrated GaInN LEDs with a six-layer graded-refractive-index anti-reflection coating made of ITO. It enhanced light-extraction efficiency by virtual elimination of Fresnel reflection.

Fourthly and lastly, as previously stated in Section 1 of this paper, the first successful and commercially viable white LED was based on a yellow garnet phosphor, $Y_3Al_5O_{12}$ doped with the optically active element Ce. Such YAG:Ce phosphors are characterized by: (1) an optical absorption band in the blue spectral range, (2) an emission band that reaches into the green and red part of the visible spectrum and has a peak wavelength of about 550 nm, (3) high chemical stability, (4) high stability under high optical irradiance, (5) short radiative lifetime (about 50–80 ns), and (6) tunability of the optical emission spectrum by adding Ga and Gd to the host material (YAG) to attain peak wavelengths ranging from about 525 to 585 nm. These favorable characteristics have made YAG:Ce the phosphor of choice for white LEDs. Another important milestone in this field was the development of red phosphors (optically excitable by the blue LED chip of a white LED) that can supplement the emission of a yellow phosphor, YAG:Ce. The YAG:Ce phosphor contains relatively little red emission so that the color temperature of white LEDs based on this phosphor is relatively high, typically 5 000 K to 6 500 K. Although the YAG:Ce phosphor can be chemically modified by partial substitution of Gd for Y, so that the peak emission wavelength of the phosphor shifts towards the red, such substitution is accompanied by a reduction in phosphor efficiency, particularly at high Gd contents; that is, fully substituted GdAG:Ce has a relatively low efficiency. Therefore, the development of novel red phosphors was necessary. Red phosphors with desirable characteristics include Eu-doped sulfide phosphors [99, 100] as well as Eu-doped nitride phosphors [101]. Whereas the sulfide phosphors have a relatively lower chemical stability, particularly in the presence of humidity, the nitride phosphors have good chemical stability, high efficiency, and exhibit little thermal quenching at elevated temperatures [101].

Note that a 'phosphor blend', a mixture of multiple phosphor materials, has been frequently employed during the last decade in order to enhance the color rendition and vary the color temperature of white LEDs. Common phosphors for a phosphor blend include YAG:Ce, Eu-doped silicate phosphors emitting in the green spectral range, and Eu-doped nitride phosphors emitting in the red spectral range. Additionally, the size-tunable colloidal Cd(Zn)Se quantum dots were developed as an alternative phosphors especially for display applications of white LEDs [102,103]. For high-power white LED applications, high thermal conductivity and high thermal stability are required to not deteriorate the phosphor's efficiency and to not shift phosphor's

Figure 14 Photographs of a white LED lamp covered by a transparent phosphor-in-glass disk and the lamp in operation. (after Ref. [108]).

chromaticity during operation at high currents and temperatures [104]. Transparent glass ceramic phosphors were investigated as a substitute for the organic polymer binders for resin-free, high-temperature and high-humidity resistant, and long-lifetime white LEDs [105–107]. Recently, Zhang *et al.* [108] showed that the phosphor-in-glass-based white LED exhibits not only excellent heat- and humidity-resistance characteristics, but also a high luminous efficacy of 124 lm/W with a correlated color temperature of 6 674 K and a color rendering index of 70, as shown in Fig. 14.

4. Future of White LEDs

As LED devices with higher efficiency and power capabilities have become available, new application areas are constantly emerging. Figs. 15(a) and 15(b) show the use of LEDs integrated into medical goggles worn by a surgeon during an operation [109, 110]. The LED-based light source promises substantial weight savings and fulfills the stringent requirements of high-quality color rendition required during medical operations. An animated pedestrian traffic signal is shown in Fig. 15(c). LED-based automotive headlights were first introduced by the Audi Car Company in 2004 using Lumileds Lighting's Luxeon devices [111]. The car is shown in Figs. 15(d) and 15(e).

LED replacement light bulbs were introduced in 2010 and became available in stores worldwide. There is a great variety of LED products. They encompass individual SMD LEDs (no external optical losses) and retrofit LED light-bulbs and tubes (with about 20% optical losses and about 10% electrical power supply losses). These products encompass a range of color temperatures (2 700 K to 6 500 K), a range of CRI values (general CRI, R_a, ranging from 75 to 95), and a range of system configurations (dimmable or non-dimmable; color-tunable or non-tunable). The luminous source efficacy will vary accordingly. Nevertheless, it can be assumed that future light sources (bulbs, tubes, as well as non-traditional smart sources) will have luminous efficacies exceeding 150 lm/W. Photographs of retrofit LED lamps are shown in Fig. 16 [112, 113].

There are clear advantages of LED lamps over their conventional counterparts, including:

- High luminous efficiency and, consequentially, low power consumption (67 lm/W, Philips, 2010; 94 lm/W, Philips, 2011; commercial LED light bulbs with source efficacies > 100 lm/W, by multiple manufacturers, 2015)

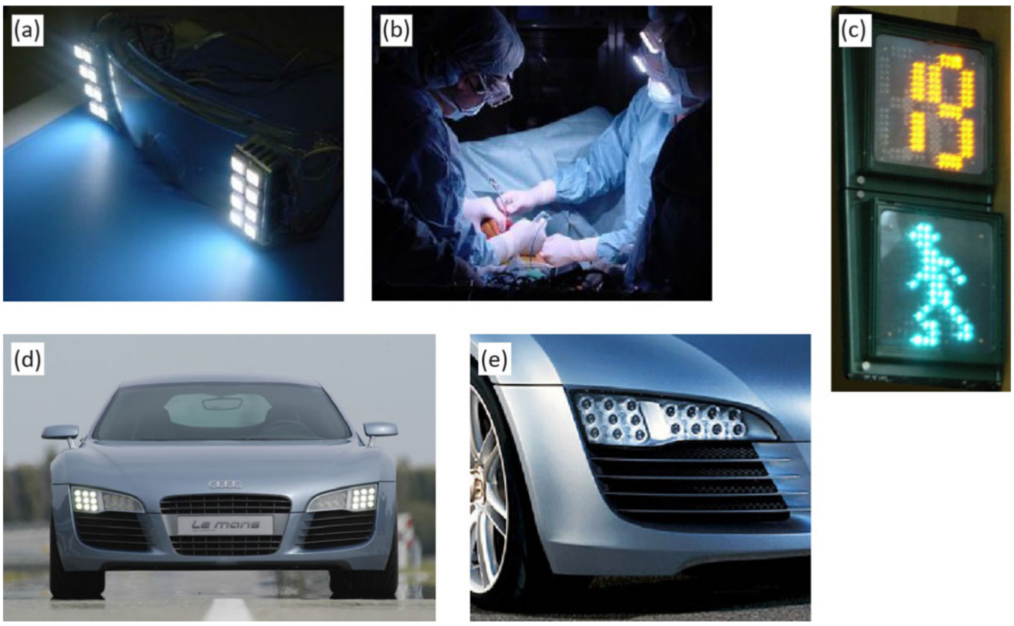


Figure 15 (a) First goggle with integrated white LEDs used for (b) illumination during medical surgery (after Shimada *et al.*, 2001; Shimada *et al.*, 2003). (c) Pedestrian sign indicating number of seconds left to cross street, located in Taipei, Taiwan (2005). (d) & (e) First automotive daytime running lights based on LEDs (adapted from Ref. [111]).

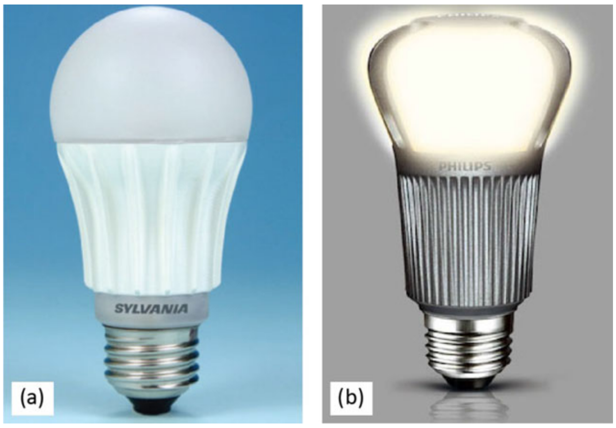


Figure 16 (a) Sylvania A19 LED lamp consuming 8W for replacement of a 40W incandescent lamp. The LED lamp offers a color-rendering index of 82 with enhanced red tone rendering and lasts 16 times longer than an incandescent lamp. The LED lamp contains no Hg and is free of UV and IR radiation minimizing discoloration and fading of materials (adapted from Ref. [110]). (b) Philips Endura LED lamp consuming 12 W and emitting a luminous flux of 806 lm for replacement of a 60W incandescent lamp (adapted from Ref. [113]).

- High quality color rendition (high color rendering index, CRI)
- Available in a wide range of correlated color temperatures (2 700 K – 6 000 K)
- No Hg (mercury) is contained in the lamp (in contrast to fluorescent lamps)
- No UV and IR radiation leading to the bleaching of fading of colored objects
- Long lifetime, e.g. 25 000 hours of operation (at 70% lumen maintenance)

These advantages make LED lamps a convincing proposition so that LED lamps are expected to displace virtually all conventional light sources. As a potential risk of LED lamps, lighting at night may disturb the body's biological clock, the circadian rhythm [114]. According to the Harvard Health Letter ("Blue light has a dark side" [115]), blue wavelengths seem to be the most harmful at night and comprise a large portion in the emission of CFLs and LED lamps. Although health issues caused by LED lamps are still under investigation, a positive side of LED lamps is their adjustability of color qualities in order to reduce or suppress any harmful effect, as compared to conventional non-adjustable lighting sources.

Furthermore, another dimension of solid-state light sources was pointed out by Schubert and Kim [116], in an article entitled: "Solid-state light sources getting smart". The authors showed that solid-state sources can be controlled (by either design or by means of real-time control), much more so than conventional light sources (i.e. incandescent and fluorescent sources). The specific parameters that can be controlled in LEDs are the (i) emission spectrum, (ii) polarization direction, (iii) color temperature, (iv) spatial emission pattern, and (v) temporal modulability. This opens up the possibility for solid-state light sources to be tailored for specific applications, and for the possibility to establish new functionalities of solid-state sources, so that they can serve for new, useful purposes. The controllability of LEDs along with their lower energy consumption and their positive environmental effects are the key advantages of solid-state sources. Next, three potential applications that show future prospects of the white LEDs will be discussed.

First, although the communication of information by means of lighting sources has not been an option for conventional lighting sources, it is indeed an option for solid-state sources. As a future prospect of white LEDs, visible light is a region of the electromagnetic spectrum which a new generation of lighting devices can use for communication. The rapid adoption of indoor and outdoor solid-state lighting will serve as a powerful platform for a new means of delivering data, visible light communication (VLC) or Li-Fi [117–119]. Conceptually, the operation mechanism of VLC or Li-Fi is reminiscent of a signal lantern. Data via a power line is piped to an LED lighting system equipped with a signal-processing system. Light shining directionally onto users and their electronic devices is able to stream data by intensity modulation so rapidly that the human eye does not recognize it. An optical sensor on a user's device detects the fluctuations in light intensity and converts them into a digital signal. Recently, indoor positioning appeared in first commercial applications, i.e., a system that connects in-store LED lights with consumers' smart phones. Shoppers are provided special deals based on their location within a store. People are able to locate items on their shopping lists or get coupons as they pass products on the aisles. Retailers can send targeted information such as recipes and coupons to consumers based on their precise location within stores [120]. Further applications of Li-Fi may include (i) LED headlights and tail lights on automobiles which let vehicles communicate with each other, (ii) traffic signals to reduce red-light violations, collisions, and fatalities, and (iii) healthcare applications. Since sensitive medical equipment could interfere with signals from cellphones and Wi-Fi, Li-Fi can provide secure and localized communications with patients, medical staff, and visitors, while lower-data-rate light-based systems can track positions of people and moveable equipment in a hospital. Global VLC/Li-Fi technology market was expected to reach $6 138 million by 2018 with an estimated CAGR of 82% from 2013 [121]. One of the important technical hurdles of this technology is upstream communication. Considering that Wi-Fi networks are bidirectional, whereas Li-Fi allows only for unidirectional streaming of data to mobile devices, getting data from those devices back to the network will be an issue to be solved [122].

Second, plant-growth factory may become a big market for LED lighting. A plant-growth factory is a closed plant-growing system that enables one to produce crops through planning and management similar to industrial products. This can be accomplished by artificially controlling environmental conditions such as light, temperature, humidity, carbon dioxide concentration, and nutrients. Lighting is a most important variable in such a plant-growth factory; lighting should be accurately controlled thereby ushering in a new era in the application of LED lighting. Although LEDs were first used to grow plants in outer space by NASA in the 1960s [123], little attention has been given to applying LEDs in agriculture until the availability of the full spectral range of colored LEDs. Unlike conventional lighting that emits in a broad wavelength range, LED lighting has the capability of growing plants with computer-controlled accuracy by, for example, emitting enhanced blue or red wavelengths that are known to effect plants and insects. Plant tissue performing photosynthesis typically absorbs mid-blue and red colors but reflect green colors. Given the advantages of LEDs (i.e. the full controllability of time, intensity, and spectrum), LED lighting makes it possible to achieve maximum efficiency of photosynthesis. LED lighting for environmentally friendly agriculture is not just a temporary trend but may become essential for survival as the world is facing global food, energy, and environmental challenges.

Third and last, another potential application of LEDs is in phototherapy. Phototherapy means treatment of patients' skin with light. Treating skin diseases with ultraviolet-B (UVB) radiation is a common type of phototherapy based on the bactericidal properties of UVB radiation. Meanwhile visible light has also been studied to directly treat dermatitis and muscle analgesia, as well as to remove bacteria in vitro [124–126]. Recent studies reported that blue LEDs, via generation of intracellular reactive oxygen species, are more effective than quartz tungsten halogen (QTH) lamps in inhibiting the proliferation of human gingival fibroblast cells [127]. Especially, a recent study of the phototoxic and bactericidal effect of blue LED irradiation revealed that LED irradiation induces apoptosis by activating a mitochondria-mediated pathway and reducing the initial growth rate of melanoma cells, which indicates that the potential of LEDs will be far greater than expected [128,129]. Oh *et al.* [130] observed that irradiation with blue LEDs reduced cell viability and thus induced apoptotic cell death with the mouse A20 and human RAMOS B-cell lymphoma cells, as shown in Fig. 17. While further research on the mechanism of autophagy in blue LED irradiation-induced apoptosis in lymphoid cells is necessary, there are certainly many potential opportunities for LEDs in new medical applications.

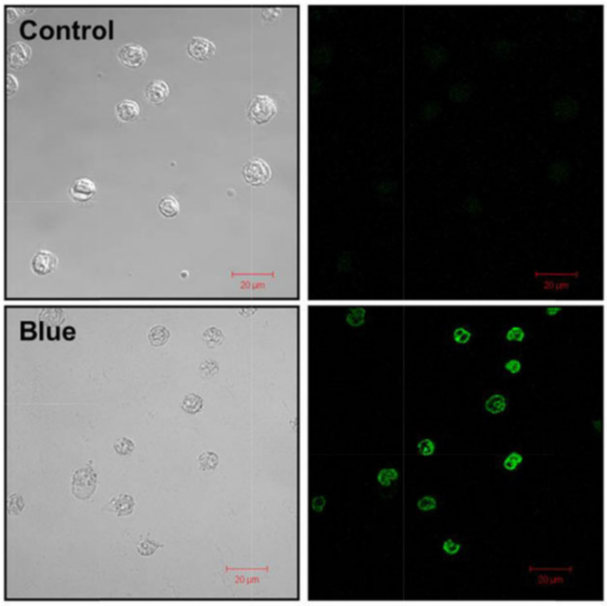


Figure 17 An apoptosis observation by an exposure to blue LEDs. The mouse A20 B-cell lymphoma cells were exposed to blue LEDs for up to 4 hours and viability was measured. A20 cells were treated with the TdT enzyme and stained with dUTP-fluorescein isothiocyanate using a TUNEL staining kit (Takara Inc., Japan). Numerous cells were stained green, indicating that apoptotic cell death occurred in the A20 cells exposed to blue LEDs (adapted from Ref. [130]).

5. Conclusion

We reviewed the history of the conception, improvement, and commercialization of the white LED. The history is testimony of diligent research and development, of successes and failures, and of researchers who made numerous attempts to create a new viable light source based on semiconductors. Prior to the advent of the white LED in 1996, conventional thinking envisioned a white LED-based light source consisting of three LED chips emitting the primary colors red, green, and blue (RGB), with each LED chip having its own power supply. However, the phosphor-based approach, based on a single blue LED chip and a phosphor that partially absorbs the blue light and reemits it as a broad green-to-red band, provided simplicity through a single power supply and, ultimately, high efficiency. The resulting white LED is a light source that is far better and more powerful than any of its conventional incandescent and Hg-discharge-based fluorescent predecessors. Whereas early white LEDs, commercially available in 1996, had a luminous efficacy of typically 5 lm/W, tremendous progress has been made as demonstrated by white LEDs with a greater-than 200 lm/W efficacy, which were commercially available in 2016. This remarkable progress, made during the last 20 years, has enabled white LEDs to far exceed the efficiency of traditional lighting devices (incandescent and Hg-discharge-based fluorescent lighting devices) thereby relegating these traditional lighting devices to a few niche applications. The progress made in white LEDs is based on advances made in blue LED chips, device architecture, phosphors, and light extraction. By any measure, the future of white LEDs looks bright and is flourishing, considering new and emerging and high-impact applications. Smart, adaptive, communicative, and tunable lighting sources have become a reality and will increasingly enable lighting technologies that are pleasant, healthy, and energy efficient.

Supporting Information

Additional supporting information may be found in the online version of this article at the publisher's website.

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