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Colour optimization of phosphor-converted flexible nitride nanowire white light emitting diodes

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Abstract

We demonstrate flexible nanowire white light-emitting-diodes (LEDs) with an optimized colour quality. The devices consist of flexible InGaN/GaN nanowire LEDs acting as pumps, capped with removable phosphor-doped polydimethylsiloxane membranes. Five different phosphors with tens of microns in grain size emitting from green to orange are investigated using both violet-blue and a blue-green nanowire-based LED pumps. In addition, a flexible nanowire white LED with a warm white emission is demonstrated using two layers of different phosphors. Compared to the previous realizations of flexible nanowire white LEDs, these novel LEDs improve the colour rendering index from 54 to 86 and show a colour tuneable from a bluish cool white colour to natural white and finally to warm white. The flexibility tests show that the LEDs can be bent down to 1.5 cm curvature radius without significant degradation. Therefore, the replacement of the nano-phosphors used in the previous realization by relatively inexpensive micro-phosphors does not degrade the good mechanical flexibility of the white nanowire LEDs.

1. Introduction

Flexible optoelectronic devices are attracting a lot of research interest, both in academy and industry [1–4]. The development of flexible light sources is one of the most rapidly evolving fields. Today, organic light emitting diodes (OLEDs) are the dominating technology for flexible light sources and have been commercialized in curved displays [5, 6]. However, in comparison to the inorganic nitride light-emitting-diodes (LEDs), the OLEDs suffer from a lower luminance and poorer stability under high current and shorter lifetime, especially in the short wavelength spectral range [7–9].

Thin-film nitride semiconductors are largely used to fabricate efficient white LEDs by converting the blue emission from an InGaN blue LED chip with yellow phosphor coverage [10, 11] to achieve a broad emitted spectrum. Thanks to their enormous contribution to energy saving, nitride LEDs have been the most important materials for solid-state lighting for the last decade. However, the mechanical stiffness of the nitride epitaxial layer and their growth on thick substrates limits their application for flexible devices. One method to fabricate nitride-based flexible sources is to perform a micro-transfer printing of LEDs to a flexible host substrate [12]. Miro-transfer printing usually uses micro-patterned elastomer stamps to deterministically transfer the micro-structured thin-film LEDs from their native substrates onto flexible holders by adjusting the rate-sensitive adhesion between the solid objects and the stamp [13]. However, this technique suffers from the high cost
related to the layer lift-off and micro-structuring. An alternative method is to use bottom-up three-dimensional nanowires (NWs) [14–19].

Thanks to their excellent material quality, nitride NWs show outstanding optoelectronic properties [20]. In addition, a core/shell geometry allows significant increases in the active area and for the design of the active region on non-polar planes [21]. In 2016, a research group, Osram, demonstrated rigid nanowire white LEDs [22] by filling a mixture of the yellow and red nano-phosphors into the air gaps between the InGaN NWs producing blue electroluminescence (EL) to enable the blue-to-white conversion.

NWs provide the possibility to fabricate flexible devices, thanks to their small footprint compared to the bending radii of the macroscopic devices [23]. In 2016, we demonstrated the first flexible white LED based on InGaN/GaN core–shell NWs and yellow nano-phosphors [18]. This white LED consisted of an NW array encapsulated in a polydimethylsiloxane (PDMS) membrane with nano-phosphors added in the gaps between the InGaN NWs and on top of the membrane. However, this first-generation flexible white LED had a poor white colour spectrum: it emitted a bluish cool white light with a high correlated colour temperature (CCT) of 6306 K and a low colour rendering index (CRI) of 54. The purpose of the present study is to demonstrate flexible white NW LEDs with optimized colour characteristics. By varying the pump wavelength and analysing five different phosphors, the CRI improved from 54 to 86 and the colour was tuned to the natural white and warm white, respectively. These results demonstrate applications of flexible white LEDs for various lighting needs. Furthermore, we show that the nanometric size of the nano-phosphors is not necessary to enable the mechanical flexibility of the white LEDs. The micro-phosphors with a grain size of several tens of microns, which are relatively easy to synthesize and have a high luminescence quantum yield [24], are suitable to fabricate the flexible NW white LEDs as will be shown in the following text.

2. Experimental method

Self-assembled GaN NWs with radial InGaN/GaN multiple quantum wells (MQWs) are used to fabricate the flexible LEDs pumping the phosphors (further referred to as ‘LED pump’) to achieve a broad emission spectrum. The NW arrays are grown on sapphire substrates by metal–organic vapour phase epitaxy (MOVPE). The morphology of the NW array is shown in the SEM image in figure 1(a) with the core–shell structure schematically illustrated in figure 1(b). The NWs are oriented along the c-axis and have the form of hexagonal prisms with diameters varying from 500 nm to 1.5 μm, and lengths of around 30 μm. Every NW is composed of an Si-doped n-GaN core grown at 1040 °C, which is separated into a heavily Si-doped bottom, partly due to the use of a high silane flux (150 nmol min−1) to promote vertical growth [25] and a lightly Si-doped upper part coming from the residual Si species. Then, the core/shell heterostructures are grown at a lower temperature that consists of the active region of seven InGaN/GaN MQWs covered with an Mg-doped p-GaN shell. The emission colour of the LED can be tuned by varying the growth temperature of the InGaN MQWs leading to a different In concentration, keeping constant the temperature at 885 °C to grow GaN barriers [26]. For this study, two samples with different targeted In content were grown using the MQW growth temperatures of 750 °C (emission in the range of 400–450 nm) and 680 °C (emission in the range of 450–500 nm). The p-GaN shell is grown at 920 °C followed by a dopant activation annealing at 700 °C during 20 min.

To investigate the impact of the pumping wavelength on the white light colour quality, two flexible LED pumps with an emitting surface size of ~1 cm2 were fabricated from the samples with the different In content. First, the NW base parts were protected with a resist layer, followed by a thin layer of Ni/Au (4 nm/4 nm) deposited on the p-GaN shells of NWs and annealed at 400 °C in an oxygen-containing atmosphere to achieve a good ohmic contact [23]. Next, the NW arrays were encapsulated in a PDMS layer, which was then mechanically peeled-off to form a suspended membrane. Contrary to the first-generation LED [18], no phosphors were added in the PDMS between the NWs in order to be able to use the same pump LED membrane with different phosphors. Afterwards, Ti/Al/Ti/Au was deposited on the n-doped base parts of the NWs. Finally, the front surface was spin-coated with a suspension of silver NWs to contact the p-GaN shells of the NW LEDs. This procedure was applied to form two flexible LEDs, which will be referred to as a ‘violet-blue’ (VB) pump and a ‘blue-green’ (BG) pump. To fabricate the flexible white LED, the phosphor powders were mixed into the PDMS with a mass ratio of 1:10 to form 300 μm-thick PDMS membranes. These membranes were applied on the top surface of the pump LEDs to convert the emission from the LED pump into the white emission spectrum.

Figure 1(c) illustrates the major steps in the fabrication process. Note that in these second-generation white LEDs, the free-standing phosphor-doped PDMS membrane was made reusable. Therefore, the same pump LED could be used for all measurements by changing the phosphor-doped capping layer. In this way, any colour variations arising from the inhomogeneities of nanowire arrays could be avoided and the impact of the phosphors on the colour performance could be directly assessed.
In total, five different phosphors mixed in free standing PDMS membranes were investigated. Table 1 shows the parameters of different phosphors. Each membrane is sequentially mounted on the two flexible pumping LEDs (one VB pump and one BG pump) to obtain different white LEDs. In addition, a flexible white LED (referred to as a third-generation LED) was obtained by stacking two layers of PDMS doped, respectively, with a yellow phosphor and with an orange phosphor in order to enhance the long wavelength component of the emission.

We performed the mechanical flexibility tests and measured the EL spectra of all the second- and third-generation white LEDs and estimated the external quantum efficiency (EQE) and wall plug efficiency (WPE) of the third-generation white LED. The CCTs and the CRIs corresponding to these EL spectra were calculated to assess the white colour quality.

Table 1. Parameters of the phosphors.

| Phosphor | Emission colour | Material | Grain size |
|----------|-----------------|----------|-----------|
| Y1       | Yellow          | Y2.94Ce0.06Al5O12 | 500 nm |
| Y2       | Yellow          | Y2.94Ce0.06Al5O12 | 5–10 μm |
| Y3       | Yellow          | Y2.91Ce0.06Gd0.03Al5O12 | 5–20 μm |
| G1       | Green           | (SrBa)1.9Eu0.1SiO4 | 2–20 μm |
| O1       | Orange          | Sr1.95Eu1.00Tb0.11Si2N8 | 5–10 μm |

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3. Results and discussion

3.1. Phosphor properties

The photoluminescence excitation and emission spectra of the five phosphors mixed in the PDMS are shown in figure 2(b). The phosphors entitled Y1, Y2 and Y3 are the well-known Ce$^{3+}$-doped garnet ($\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$) phosphors. Note that Y1 is the same nano-phosphor that was previously used for the first-generation LED [18] with a particle size of only 500 nm. The other four phosphors used in this study have a larger grain size ranging from 2–20 $\mu$m. The excitation bands of Y1 and Y2 are centred at $\sim$465 nm, which corresponds to the first allowed $4f$–$5d$ transition of Ce$^{3+}$ ions, from the Ce$^{3+}$ $4f$ level to its lowest $5d$ ($^2A_{1g}$) level. The secondary excitation peak at 340 nm is attributed to the second allowed $4f$–$5d$ transition, $4f$($^2F_7/2$)/$5d$($^2B_{1g}$), of Ce$^{3+}$ ions [27].

The YAG:Ce phosphors lack the red spectral component, compared to the natural solar spectrum. One method to boost the red emission is to substitute Y$^{3+}$ with larger ions (e.g. Tb$^{3+}$, La$^{3+}$ and Gd$^{3+}$) to increase the crystal field splitting and to tune the Ce$^{3+}$ emission to a longer wavelength [29]. For the phosphor Y3, the doping with Gd$^{3+}$ leads to a redshift of the central emission wavelength to 556 nm, compared with $\sim$547 nm in Y1 and Y2, as shown in figure 2(b). The Gd$^{3+}$ doping also results in a flat and broad excitation spectrum from 300–460 nm. The redshift of the emission band for Y3 in comparison with Y1 and Y2 makes it more appropriate for producing white light with a warmer white colour and a higher CRI.

One of the widely used green phosphors compatible with blue pumping is an Eu$^{2+}$-activated ($\text{Sr, Ba}_2\text{SiO}_4$) orthosilicate. This system can be excited with a wide range of wavelengths from near-UV to blue (from 380 nm to 475 nm), and shows intense green or yellow emissions with the peak wavelength ranging from 490–580 nm. The emission colour depends on the Sr to Ba ratio as the Eu$^{2+}$ emission is effectively influenced by the

![Figure 2.](image-url)
surrounding crystal environment and crystal field. In Ba$^{2+}$ sites (in Sr$^{2+}$ sites), the doped Eu$^{2+}$ ions usually exhibit green emission (yellow-green emission), respectively. Furthermore, this intermediate (Sr,Ba)$_2$SiO$_4$: Eu$^{2+}$ composition presents a better photoluminescence stability at elevated temperatures [30]. In this investigation, we use (Sr,Ba)$_2$SiO$_4$: Eu$^{2+}$ phosphor G1 showing a broad green emission peaked at around 509 nm under a 473 nm blue excitation. The emission arises from the 4f$^6$5d$^1$-4f$^7$ transitions of Eu$^{2+}$ ions [31].

A nitride-based Sr$_2$Si$_5$N$_6$: Eu$^{2+}$ system is well known for exhibiting long-wavelength orange-red emission bands upon UV and blue excitations. This phosphor system is excitable in a wide range of wavelengths from 360–470 nm. It is characterized by an excellent thermal stability at elevated operating temperatures because of its robust host covalency of N$^{3−}$ [32]. The orange phosphor O1 used in this investigation is an Sr$_2$Si$_5$N$_6$: Eu$^{2+}$ compound, which shows an orange emission ranging from 550–700 nm and peaked at around 612 nm under the blue excitation with 451 nm (figure 2(b)), due to the spin-allowed 4f$^6$5d$^1$-4f$^7$ transitions of Eu$^{2+}$ ions [33]. In this investigation, Sr$_2$Si$_5$N$_6$: Eu$^{2+}$ was used alone, and also in combination with a yellow phosphor to demonstrate a suitable white LED system with a warmer white colour.

All the above-described phosphors are widely used for white LEDs due to their high photoluminescence efficiency and adequate thermal stability.

### 3.2. Flexible LED pumps

Two different pumping LEDs were used to excite the phosphor-doped PDMS membranes. The EL spectra of these two LEDs are shown in figure 2(a) for an injection current of 300 mA. The LED, referred to as ‘VB’, has a violet emission peak at 404 nm and a broad blue shoulder centred at 464 nm. By comparison with the excitation spectra of the phosphors traced in figure 2(b), the blue shoulder overlaps very well with the excitation spectra of all the five phosphors. However, the violet peak overlaps only with the excitation spectra of phosphors Y3, O1, and G1, but it is mismatched with respect to those of Y1 and Y2. This may lead to an insufficient conversion of the violet light in the resulting spectrum of the white LED, emitting a cool white colour. The EL emission of the LED referred to as ‘BG’ consists of one blue peak at 439 nm and one green peak at 485 nm. The spectrum of the LED BG shows a good overlap with the excitation spectra of the phosphors Y1, Y2, G1 and O1 and a partial overlap of the blue component for the phosphor Y3.

### 3.3. White flexible LEDs

Figure 3 displays the EL spectra of the second-generation flexible white LEDs measured with two different LED pumps and five different phosphors mixed in 300 μm-thick PDMS membranes. As expected, all the white LEDs show the EL spectra with a long-wavelength component (figure 3), which is in good agreement with the emission spectra of the phosphors traced in figure 2. Figure 3(a) shows the EL spectra of the white LEDs pumped with the LED VB. The violet peak of the LED pump is partially absorbed by all the phosphors, but remains present in the resulting EL spectra. The flexible white LEDs with Y1 and Y2 exhibit similar EL spectra. This is indeed expected since the Y1 and Y2 phosphors differ only by the grain size. The blue shoulder is almost entirely converted to the long-wavelength emission thanks to the good spectral overlap with the excitation spectra of the phosphors. This can be well observed in figure 4(a), which shows the details of the conversion process for one specific example. It compares the excitation and emission spectra of the phosphor Y2 with the EL spectrum of the pump LED VB, and the resulting EL spectrum of the white LED. No blue shoulder can be seen in the final spectrum of the white LED. The blue component is also strongly absorbed by G1. However, for the phosphors Y3 and O1, the blue shoulder markedly remains in the EL spectra of the white LEDs.

Figure 3(b) shows the EL spectra of the white LEDs pumped with the LED BG. The phosphors Y1, Y2 and G1 exhibit a strong absorption of the blue component, while the phosphors Y3 and O1 only partially absorb the blue peak of the pumping LED. This result is consistent with the spectra obtained for the LED VB showing the nearly total absorption of the blue shoulder for Y1, Y2 and G1, but presenting a residue of this shoulder for the phosphors Y3 and O1, as discussed earlier. For the green peak at 485 nm of the LED BG pump, the white spectra for all the phosphors show an important unconverted residual peak. In particular, for G1, the green emission from the LED pump merges with the converted green emission of the phosphor, forming a broad green peak centred at 509 nm.

### 3.4. High-colour quality white flexible LED with a warm white colour

From the EL spectra of the white LEDs shown in figure 3, it is seen that the most wide-spread yellow phosphors, i.e., Y1, Y2, and Y3, lack the emission at wavelengths longer than 600 nm. This results in a rather poor colour quality of obtained white light. In order to demonstrate a white LED with a broad spectrum covering the whole visible spectral range, we fabricated the third generation of flexible white LEDs by stacking a PDMS membrane doped by the orange phosphor O1 onto a membrane doped with the yellow phosphor Y2. The stack of the two membranes is pumped by the LED VB. The EL spectrum of this third-generation LED is traced as the green solid
This figure also displays the excitation and emission spectra of the phosphor O1 and the EL spectrum of the second-generation white LED consisting of an LED VB pumping the Y2 doped membrane. We can observe that the violet residual emission from the pumping LED and the yellow component corresponding to the phosphor Y2 in the second-generation LED are both partially converted to orange by the phosphor O1.

In order to estimate the EQEs and WPEs, we measured the emitting powers of the third-generation white LED and the pump LED VB using a power-meter with a large area sensor having a calibrated flat spectral response in the visible range. Taking into account the size of the LED and of the sensor, only the emission with a light cone angle of 75° is detected with this set-up geometry, which leads to an underestimation of the total emitted power [18]. The EQE is defined as the ratio between the photon number emitted by the LED and the number of injected electrons. The WPE is given by the ratio between the optical power of the LED and the injected electrical power. At 300 mA, the values of the measured EQE and WPE of the third-generation white LED are 0.63% and 0.18%, respectively. These low values mainly originate from the low performance of the LED pump built from self-assembled NWs, which exhibit strong compositional and morphological inhomogeneities [18, 19]. Indeed, at the same injected current, the pump LED VB has a measured EQE of 2.63% and WPE of 1.04%. Note that the EQE and WPE values are underestimated because of the detection system. By taking the ratio between the WPE values of the third-generation white LED and of the pump LED VB, the efficiency of the downconversion by the two layers of phosphors can be estimated at around 17%.

3.5. Colour quality parameters

Table 2 shows the commission internationale de l’éclairage (CIE) coordinates (CC), CCT and CRI of the white LEDs with different phosphors and different LED pumps. The locations in the CIE 1931 colour chart of all the tested LEDs are shown in figure 5. One important target of this study is to improve the light quality from the values previously obtained for the first-generation white LED (i.e. cool white colour with poor CRI) to get closer to the natural sunlight (CCT = 4000–5000 K and CRI = 100), which is also a target for most lighting industrial applications. The CCTs of the white LEDs with the pumping LED VB are ~4150 K for Y1 and Y2, corresponding to a natural white colour. Note that, compared with the first-generation flexible bluish cool white LED reported in [18], the white colour became warmer by doubling the phosphor density (phosphor-to-PDMS mass ratio from 1:20 to 1:10) [18] in the membrane: the CCT shifted from 6306 K to 4160 K. This improvement is due to the stronger conversion of the phosphor, which leads to a lower residual emission from the pump and a more important component in the long-wavelength range in the final white LED spectrum. We also conclude that it is...
not necessary to fill the gaps between the nanowires with phosphor grains—efficient conversion can be achieved by capping the top surface of the LED. The CCTs for Y1 and Y2 with the pumping LED BG are more than 2000 K higher than those with the LED VB. This higher CCT corresponding to a cool white colour is due to the poorly converted green peak around 480 nm in the pump spectrum. This explanation is also applicable to the other phosphors. It clarifies why the emissions of white LEDs with the pump LED BG (red dots in figure 5) are located toward a higher CCT in the colour chart than those with the pump LED VB (blue dots in figure 5). The CRIs with

**Table 2. CC, CCT, and CRI of the white LEDs with different phosphors and different LED pumps.**

| Phosphor | LED pump | CC (x, y) | CCT (K) | CRI |
|----------|----------|-----------|---------|-----|
| Y1       | LED VB   | (0.3877, 0.4306) | 4160 | 76  |
|          | LED BG   | (0.3121, 0.3912) | 6209 | 82  |
| Y2       | LED VB   | (0.3852, 0.4180) | 4147 | 70  |
|          | LED BG   | (0.3038, 0.3657) | 6665 | 86  |
| Y3       | LED VB   | (0.3450, 0.4012) | 5134 | 71  |
|          | LED BG   | (0.2035, 0.2488) | >100 000 | NA |
| G1       | LED VB   | (0.2751, 0.3924) | 7641 | 71  |
|          | LED BG   | (0.1932, 0.2965) | NA   | NA  |
| O1       | LED VB   | (0.4065, 0.2687) | 2166 | 62  |
|          | LED BG   | (0.2773, 0.2389) | 18846 | 55 |
| O1 on Y2 | LED VB   | (0.4268, 0.3824) | 2993 | 85  |

not necessary to fill the gaps between the nanowires with phosphor grains—efficient conversion can be achieved by capping the top surface of the LED. The CCTs for Y1 and Y2 with the pumping LED BG are more than 2000 K higher than those with the LED VB. This higher CCT corresponding to a cool white colour is due to the poorly converted green peak around 480 nm in the pump spectrum. This explanation is also applicable to the other phosphors. It clarifies why the emissions of white LEDs with the pump LED BG (red dots in figure 5) are located toward a higher CCT in the colour chart than those with the pump LED VB (blue dots in figure 5). The CRIs with
the LED BG are higher than those with the LED VB. The flexible white LED only with Y2 pumped by the LED BG has the best CRI value of all the tested devices (CRI = 86), which is comparable with the commercial white rigid LEDs. For the phosphors Y3, G1 and O1, with the pumping LED BG, the white LEDs have unbalanced strong components in the short-wavelength range so that their CCs are shifted from the centre of the white colour zone in the CIE colour chart (see figure 5). With the LED VB pump, the CCT decreases from 7641 K to 5134 K and to 2166 K for the phosphors G1, Y3, and O1, respectively. This is due to the increased emission wavelength of the phosphors, corresponding to the colours of green, yellow and orange for G1, Y3, and O1, respectively.

For the third-generation flexible white LED, which combines Y2 and O1 (blue circle in figure 5), compared to the second-generation white LEDs pumped with LED VB and having only one-layer of phosphor Y2, the colour shifts from natural white (CCT = 4147 K) to warm white (CCT = 2993 K). This decrease of the CCT is indeed expected, thanks to the richness of the long-wavelength component emitted by the additional O1. The broad spectrum of the white LED with this phosphor mixture also results in a high CRI of 85.

These second- and third-generation white LEDs cover the warm white, natural white and cool white, corresponding to the CCTs varying from 2166–6665 K, and several of them have a high CRI.

3.6. Mechanical flexibility

The mechanical flexibility of the second- and third-generation white LEDs was tested by comparing the I–V curves and the EL spectra before and after 50 repeatable bendings for different curvature radii down to 1.5 cm. Photos of the third-generation NW white LED under operation in a flat condition and with a bending radius of 1.5 cm are shown in figures 4(c), (d). All the flexible LEDs did not show any emission or spectral degradation after the bending tests. The devices have a good mechanical flexibility, even for the LEDs with the phosphors with a relatively large grain size, i.e., Y2, Y3, G1, and O1. In the tested range of the curvature radii, their behaviour is comparable to the first-generation LEDs fabricated with the nano-phosphors additionally filling the gaps between the nanowire arrays. This means that the nanometric size of the phosphor grains is not mandatory to fabricate the flexible LEDs for applications requiring bending curvature radii in a centimetre range and the standard micro-phosphors can ensure the mechanical flexibility in these conditions. Nowadays the nano-phosphors are mostly used for the biomedical applications where the shape and the nanometric size plays an important role [34]. However, it is still challenging for nano-phosphors to compete with the standard solutions in terms of the quantum efficiency. For commercial lighting applications, the luminescent quantum efficiency and the yield of synthesis are more important criteria than the grain size and shape. Therefore, the micro-phosphors, which are less challenging to synthesize than the nano-phosphors [24], are more suitable for the flexible lighting application requiring a moderate bending.
4. Conclusion

We demonstrated that the colour performance of flexible white NW LEDs can be adjusted by changing the pumping LEDs and the phosphors. The use of phosphor with a submicrometric grain size is not mandatory to maintain the mechanical flexibility of the white NW LED. The device configuration, where a phosphor-doped PDMS membrane is mounted on top of the LED pump instead of phosphors being infiltrated between the nanowires, shows an efficient blue to white conversion. The replacement of the nano-phosphors by micro-phosphors circumvents the challenging procedure of the synthesis of the efficient phosphors with nanoscale grains. Compared to the first-generation flexible white NW LED, the second-generation LEDs using a single phosphor layer show an improvement of the CRI from 54–86 and tune the bluish cool white colour to natural white or warm white. The third-generation flexible white NW LED, obtained by stacking two layers of yellow and orange phosphors, emits a warm white light and has a higher CRI of 85, which is higher than the white LED with the same pump but with only one layer of yellow phosphor. These high-CRI flexible LEDs with different CCTs can be used for different lighting applications, e.g. for extended light sources integrated on non-rectilinear surfaces for ambient lighting. By using different NW LED pumps and changing phosphors, the colour quality can be adjusted to fit various applications, e.g., the high-CRI LED emitting a cosy warm white light (achieved with the third-generation LED) and the LED emitting natural white light with a good CRI (achieved for example with the second-generation LED with a phosphor Y1 pumped by an LED VB).

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