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Enhanced bandwidth of white light communication using nanomaterial phosphors

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Abstract

The bandwidth of white light emitting diodes (WLEDs) is an important factor that affects most of the system performances in visible light communication (VLC). It is mainly limited by the down-conversion phosphors. We propose in this paper to employ nanomaterial phosphors with short fluorescence lifetime and high quantum yield in VLC. The white-emitting device of bandwidth-based lifetime was fabricated by using several kinds of nanophosphors with different fluorescence lifetimes. Moreover, we proposed two theoretical models to analyze the factors that affect bandwidth. Compared with the commercial YAG-based WLEDs, the bandwidth of nanophosphor-based WLEDs can be improved over three times and close to the blue excitation sources. Our study indicates that nanophosphors can become promising fluorescent materials in VLC, and provides a new direction for developing wide-bandwidth VLC systems.

Keywords

nanomaterial; visible light communication; white light emitting diodes

1. Introduction

Visible light communication (VLC) offers a new technology to augment the existing radio frequency approaches (such as Wi-Fi) and to satisfy the growing demand for wireless data communication due to its low electromagnetic pollution, high confidentiality and large capacity [1–5]. Illumination and data communications become available at the same time by using white light emitting diodes (WLEDs) [6–10]. Most of the important performances of VLC rely on the modulation bandwidth of WLEDs including bit error rate [11, 12], the rate and the capacity of communication [13–16].

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Effective lighting devices can be realized by combining phosphor materials with blue chips [17–21]. The commercial phosphors with long fluorescent lifetime limit the modulation frequency, such as Yttrium aluminum garnet (YAG) [22, 23]. Thus, only a few megahertz (MHz) was realized for these conventional phosphor-based LEDs. Therefore, the nanomaterials with short relaxation time and high quantum yield (QY) have attracted extensive attention [24, 25].

In recent years, quantum dots (QDs) exhibit unique characteristics of tunable band gap, broad absorption, and photo stability, making them highly attractive materials for a wide range of applications [26–30]. Research regarding QDs as color conversion in WLEDs has been particularly high-lighted, because of their tunable emission over the entire visible region, high QY, and facile fabrication [31–38].

We explored the nanomaterials as the down-conversion phosphors for the fabrication of WLEDs in VLC and proposed two theoretical models to analyze the factors of affecting bandwidth from the principle of white light generation. Three kinds of nanomaterials with different fluorescence lifetimes were employed to analyze the lifetime-dependent modulation frequency, and we demonstrated their potential for VLC through achieving a bandwidth over three times wider than conventional phosphors.

2. Methods

2.1. Materials

Dodecanethiol (DDT, 98%), tributylphosphine (TBP, 95%), oleylamine (OLA, 90%), PbBr_2 (99.0%) and oleylamine (80%–90%, OLA) and zinc acetate dehydrate ($\text{Zn}(\text{OAc})_2$, ... $\geq 97\%$) were purchased from Aladdin. Acetone, methanol, hexane, toluene, poly(styrene-co-maleic anhydride) (PSMA, $M_w \approx 1700$, styrene content 68%), anhydrous tetrahydrofuran (THF, 99.9%), cadmium oxide (CdO , 99.99%), trioctylphosphine (TOP, 90%), oleic acid (OA, 90%), and 1-octadecene (ODE, 90%) were bought from Sigma- Aldrich. Cs_2CO_3 (99.9%) was purchased from J&K. Toluene (99.5%) and Poly(vinyl pyrrolidone) (PVP, K30, $M_w \approx 40\,000$) were purchased from Beijing Chemical Reagent Company. Poly [(9,9-dioctylfluorenyl-2,7-diyl)-co-(1,4-benzo-{2,3}-thiadiazole)] (PFBT, $M_w = 100\,000$ –157 000) was purchased from ADS Dyes (Quebec, Canada). PF-DBT5 was prepared according to the method described in our previous report [39]. Selenium powder (Se, 100 mesh), and sulfur powder (S, 99.9%) were obtained from Alfa Aesar. All chemicals were used directly without further treatment. YAG phosphor was purchased from Intematix. UV glue NOA60 was obtained from LIENHE Fiber Optics.

2.2. Preparation of CdSe/ZnS QDs

According to the previous reported method (single-step synthesis) [40], the CdSe/ZnS QDs with chemical-composition gradients were prepared. As a typical synthetic procedure, 0.4 mmol of CdO , 4 mmol of zinc acetate, 17.6 mmol of oleic acid, and 20 ml of 1-octadecene were placed in a 100 ml round flask. The mixture was heated to 150 °C, degassed under 100 mTorr pressure for 20 min, then filled with N_2 gas, and further heated to 310 °C to form a clear solution of $\text{Cd}(\text{OA})_2$ and $\text{Zn}(\text{OA})_2$. At this temperature, 0.4 mmol of Se powder and

2.9 mmol of S powder both dissolved in 3 ml of TOP were quickly injected into the reaction flask. After the injection, the temperature of the reaction flask was set to 300 °C for QD growth. The flask was then cooled to room temperature to stop the growth. QDs were purified by adding 20 ml of chloroform and 3 times that volume of acetone; the final pure QDs were dispersed in hexane.

2.3. Preparation of CsPb(Br_{0.55}I_{0.45})₃ QDs

Preparation of Cs-oleate: Cs₂CO₃ (0.8 g), OA (2.5 ml) and ODE (30 ml) were added into 100 ml 3-neck flask, degassed and dried under vacuum for 1 h at 120 °C. Then, the reaction solution was heated to 150 °C under N₂ until the solution was clear. Synthesis and purification of CsPb(Br_{0.55}I_{0.45})₃ NCs: ODE (10 ml) and PbI₂ (0.073 g), PbBr₂ (0.078 g) were loaded into a 50 ml 3-neck flask, degassed and dried by applying vacuum for 1 h at 120 °C. Dried OLA (1 ml) and dried OA (1 ml) were injected to the flask at this temperature. After the solution became clear, the temperature was raised to 180 °C and Cs-oleate solution (0.8 ml, 0.1 M in ODE, preheated to 100 °C before injection) was quickly injected. 5 s later, the reaction mixture was cooled down to room temperature by an ice-water bath. The reaction mixture was separated by centrifuging for 10 min at 5000 rpm. After centrifugation, the precipitate was redispersed in 5 ml toluene.

2.4. Preparation of PDs

The green-emitting polymer dots (G-PDs) were made from PFBT [41, 42]. The red-emitting polymer dots (R-PDs) were synthesized by PF-DBT5 and PFBT at a weight ratio of 3:5 [43]. To prepare them, corresponding polymer(s) were dispersed in 5 ml THF with amphiphilic functional PSMA (10 ppm) in a 20 ml transparent glass bottle to form a homogeneous mixture with ultrasonic treatment. The mixture was quickly injected into deionized water (10 ml) in a bath sonicator and then kept at 90 °C under nitrogen gas on a hot plate in order to remove THF. Finally, a 0.22 μm filter was used to remove any large polymer particles in the solution at room temperature.

2.5. Fabrication of WLEDs

The blue LED chips with emission wavelength centered at 460 nm were used to fabricate the WLEDs. The prepared CdSe/ZnS QDs and CsPb(Br_{0.55}I_{0.45})₃ QDs, which were dispersed in hexane, mixed with a certain amount of UV glue, respectively. The PDs and YAG were also mingled with defined amount of UV glue. Ultrasonic and vibratory treatments were used to make them homogeneous mixtures. Then, the four mixtures were added dropwise onto four blue LED chips and solidified 1 min under the ultraviolet light irradiation.

2.6. Characterizations

Photoluminescence (PL) spectra were measured using a Zolix Omni-λ300 luminescence spectrometer. The UV-vis absorption spectra were obtained by a Shimadzu TU-1810 spectrophotometer. The absolute PLQYs of the samples were obtained on a fluorescence spectrometer (FLS920P, Edinburgh Instruments) equipped with an integrating sphere. The images of the morphology of the nanomaterials were taken using a TECNAIF20 transmission electron microscope (TEM). Time-resolved PL spectra were measured by a

fluorescence spectrometer (mini- τ , Edinburgh Photonics) equipped with an EPL405 laser diode. When measured the decay curve, a 5 μ s separation was employed to avoid the PL accumulation. Optical signal to electrical signal conversion was achieved by employing a photoelectric detector BPW21 (Siemens Semiconductor Group). The modulation bandwidths of WLEDs were acquired using an Agilent HP 8753ES network analyzer (300 KHz–3 GHz).

3. Results and discussion

Highly efficient yellow emitting CdSe/ZnS QDs have been used as the color conversion materials which was synthesized by reported methods [40]. Compared with the wavelength of YAG phosphor, CdSe/ZnS QDs have narrow full width at half maximum (FWHM), nearly perfect PL and high QY (70%) that were chosen in the following section. The absorption (Abs) and PL spectra of CdSe/ZnS QDs are shown in figure 1(a). The emission peak is 570 nm, absorption peak is 550 nm and the FWHM is 34 nm. Its TEM image is shown in figure 1(b), which indicates the average particle size is 7.2 nm.

Two WLEDs were fabricated by combining a 460 nm GaN LED chip with YAG and CdSe/ZnS QDs, respectively. The flow diagram of fabricating WLEDs is shown in figure S1, which is available online at stacks.iop.org/NANO/29/455708/mmedia. Figure 2(a) shows the emission spectra of the YAG and CdSe/ZnS QD-based WLEDs.

The commercial WLEDs are generally fabricated by combining blue LED with yellow emitting YAG phosphor; however, the slow response of YAG phosphor limits the available channel bandwidth. According to previous studies, the commercial YAG-based WLEDs only have a 3 dB bandwidth about 1 MHz [44]; hence the bandwidth and the transmission data rate are also limited in VLC. In order to analyze the effects of CdSe/ZnS QDs as the color converting material for VLC, we firstly measured their response to a small signal modulation to determine their modulation bandwidth (figure 2(c)). For the bandwidth measurement, a continuous wave bias current was employed to drive the device electrically, which combined the direct current (DC) from a constant current source with the reference (RF) signal from a network analyzer (Agilent HP 8753ES). The frequency response measurements were taken at 350 mA current, while a network analyzer was used to record the frequency response, as presented in figure 2(c). The output emission of WLED was collected by a pair of reflective-concentrated cups with diameter of 20 mm and focused onto a high-speed silicon PIN photo-receiver (BPW21) with an effective active diameter of 2.73 mm. The communicating distance was approximately 10 cm.

Figure 2(b) show the comparisons of frequency responses and bandwidths among three different LED modules respectively. The maximum 3 dB bandwidth of GaN LED chip is 11.8 MHz. The corresponding maximum 3 dB bandwidths of WLED were improved from 3.6 to 9.8 MHz after YAG was replaced by CdSe/ZnS QDs, which was higher than that of the reported phosphor-based WLEDs [45, 46].

The radiative relaxation of YAG is related to the trap state, which achieve afterglow luminescence [47]. In contrary, CdSe/ZnS QDs is a typical band edge emission nanomaterial

with shorter lifetime than YAG phosphor [48]. The radiative relaxation is the key factor to affect the bandwidth.

Figure 3(a) shows the generation process of the white light with the contributions of the phosphors and the pumping source. Therefore, the modulation bandwidth of WLED is influenced by both GaN chip and the phosphor. Three steps were considered to investigate several factors on the bandwidth of WLEDs including blue emission generation, excitation of phosphor film and output of white light.

It has been demonstrated that the modulation bandwidth of blue LEDs can be improved by shorting the minority carrier lifetime in active region [49, 50]. Equation (1) shows the relationship between the modulation frequency and the minority carrier lifetimes,

$$M_r(f) = \frac{1}{\sqrt{1 + (2\pi f\tau)^2}}, \quad (1)$$

where $M_r(f)$ is the degree of modulation; $\tau^{-1} = \tau_r^{-1} + \tau_{nr}^{-1}$, τ_r and τ_{nr} are the radiative and non-radiative lifetimes, respectively. When $M_r(f)$ is 1/2, the corresponding 3 dB bandwidth is shown in equation (2),

$$f_{3dB} = \frac{\sqrt{3}}{2\pi\tau_r}. \quad (2)$$

In order to simplify the discussion, we neglected the non-radiative recombination in GaN LED active region. As shown in figure 3(b), the 3 dB bandwidth was simulated with the increase of radiative lifetimes from 1 ns to 1 μ s. It is noted that f_{3dB} is inverse proportional to τ_r . The bandwidth of GaN LED was 11.8 MHz in our experiment, therefore, the corresponding theoretical lifetime should be 23.3 ns.

According to this, we speculated that the modulation bandwidth of the color converted phosphor was also inverse proportional to the lifetime of phosphors and the theoretical model could be depicted in equation (3)

$$M_R(f) = \frac{1}{\sqrt{1 + (2\pi f\tau_r)^2}} \frac{1}{\sqrt{1 + (2\pi f\tau_R)^2}}, \quad (3)$$

where $M_R(f)$ is the degree of modulation of phosphor, and τ_R is the radiative lifetime of phosphor. According to this equation, figure 3(c) shows the 3 dB theoretical fitted curves when τ_R is 1 ns, 5 ns, 10 ns, 50 ns, respectively. The white emission eventually combines both of the blue light and the phosphor's emission. Therefore, the final theoretical model can be expressed in equation (4)

$$M_w(f) = \frac{\Phi_{B0}}{\Phi_{B0} + \Phi_{Y0}} \frac{1}{\sqrt{1 + (2\pi f\tau_r)^2}} + \frac{\Phi_{Y0}}{\Phi_{B0} + \Phi_{Y0}} \frac{1}{\sqrt{1 + (2\pi f\tau_R)^2}}, \quad (4)$$

where $M_w(f)$ is the degree of modulation of white light, the Φ_{B0} and Φ_{Y0} are the radiation fluxes of blue light and yellow light in white light, respectively. When the lifetime of the GaN chip was 23.3 ns, by varying the proportion of blue light in the whole spectra with different lifetime of phosphors, the 3 dB theoretical fitted curves are shown in figure 3(d), with $\Phi_{B0}/(\Phi_{B0} + \Phi_{Y0})$ increased, the bandwidth increased accordingly.

In order to further confirm the modeling results and to improve the modulation bandwidth of device, polymer dots (PDs) and $\text{CsPb}(\text{Br}_{0.55}\text{I}_{0.45})_3$ QDs were used as phosphors to fabricate WLEDs, respectively. The images of devices are shown in figure S2. The absorption, PL spectra and TEM images of PDs and $\text{CsPb}(\text{Br}_{0.55}\text{I}_{0.45})_3$ QDs are shown in figure S3. According to the spectra of different phosphors based WLEDs (figure 4(a)), the ratios of blue light in the whole spectrum can be calculated (yellow line:33.1%, violet line:42.3%, blue line:43.7%, cyan line:22.1%). Four fluorescent decay curves were measured (figure 4(b)). The corresponding average lifetimes are 1.5 ns for PDs, 11.2 ns for CdSe/ZnS QDs, 19.8 ns for $\text{CsPb}(\text{Br}_{0.55}\text{I}_{0.45})_3$ QDs and 68.7 ns for YAG, respectively. As shown in figure 4(c), the bandwidth of $\text{CsPb}(\text{Br}_{0.55}\text{I}_{0.45})_3$ QD-based device is 8.2 MHz, which is smaller than that CdSe/ZnS QD-based device of 9.8 MHz. The PD-based WLED possessed the highest modulation bandwidth about 11.1 MHz, which was extremely close to the blue LED bandwidth of 11.8 MHz.

Table 1 summaries the characteristics of theoretical and experimental 3 dB bandwidths and the optical properties of the as-fabricated nanophosphor WLEDs. The histograms are shown in figure S4. The experiment results were in accordance with our expected model. The WLED bandwidths not only depended on the pumping blue source, but also closely relied on the fluorescent lifetime. When the blue LED bandwidth was fixed, the bandwidth of as-fabricated WLED would be determined by the fluorescent lifetime of phosphors.

Then, we analyzed the influence of blue light component for the modulation bandwidth of WLEDs. According to equation (4), the same type of blue GaN LEDs and phosphors were used to keep τ_r and τ_R as a constant. The 3 dB theoretical fitted curve is shown in figure 5(a) (yellow line). We changed the ratio of Φ_{B0} in whole white light by controlling the concentration of CdSe/ZnS QDs, and then tested the bandwidth. As shown in figure 5(a), the measured results (cyan dot line) agreed with the theoretical model, with the increase of Φ_{B0} , the corresponding 3 dB bandwidth of device increased accordingly. Figure 5(b) shows the output spectra of CdSe/ZnS QD-based WLEDs with different ratios in the whole spectra: 29.8%, 42.3% and 58.2%, respectively. Table 2 show the theoretical and experimental bandwidth as well as CIE coordinates of CdSe/ZnS QD-based WLED when the blue light ratios were 29.8%, 42.3% and 58.2%, respectively. From their CIE 1931 chromaticity

diagram (figure S5), it is observed that the emitting light of the asfabricated devices were all in white light region.

4. Conclusions

In summary, we studied the effects of phosphors with different fluorescence lifetimes on the bandwidth of WLEDs and proposed two theoretical models to analyze the factors affecting the bandwidth. The results showed that compared with conventional phosphors, the shorter fluorescence lifetime nanophosphors increased the bandwidth from 3.6 to 11.1 MHz, which was close to the GaN excitation sources. Moreover, with the proportion of blue light in the whole white light increased, the bandwidth increased accordingly. These experimental results are consistent with our theoretical models. Our findings provide a new direction to increase the bandwidth of VLC.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Figure 1. Absorption (Abs) and photoluminescence (PL) spectra of CdSe/ZnS QDs; (b) the corresponding TEM image.

Figure 2. (a) Normalized PL spectra and (b) the output responses of GaN chip, CdSe/ZnS QDs and YAG WLEDs with a working current of 350 mA; (c) experimental setup of VLC using WLEDs.

Figure 3.

(a) Schematic graph of white light generation through phosphor-based WLEDs; (b) the variation of 3 dB bandwidth versus carrier lifetime from 1 ns to 1 μ s in GaN; (c) the variation of 3 dB bandwidth versus carrier lifetime from 1 ns to 100 ns in GaN with the change of phosphors lifetime; (d) the variations of 3 dB bandwidth with different blue ratios in the white light.

Figure 4.

(a) The output spectra of WLEDs with four phosphors including PDs, CdSe/ZnS QDs, CsPbX₃ QDs and YAG; (b) PL decay curves and (c) modulation bandwidth of four LEDs.

Figure 5.

(a) Theoretical fitted curve of 3 dB bandwidth and the measured bandwidth of CdSe/ZnS QD-based WLED; (b) output spectra of CdSe/ZnS QD-WLEDs with different ratios in the whole spectra.

Table 1.

Characteristics of the studied WLEDs.

Sample	Lifetime/ns	TBW1/MHz	TBW2/MHz	ABW/MHz	CIE(x,y)
GaN	23.3	—	—	11.8	(0.129, 0.081)
PDs	1.5	11.6	11.7	11.1	(0.332, 0.346)
CdSe/ZnS	11.2	9.2	10.1	9.8	(0.323, 0.341)
CsPb(Br _{0.55} I _{0.4})	19.8	7.3	8.6	8.2	(0.324, 0.333)
YAG	68.7	3.4	4.3	3.6	(0.347, 0.392)

Note. TBW1, TBW2 were the equation (3), equation (4) theoretical bandwidths, respectively. ABW: actual bandwidth. CIE: Commission Internationale de L'Eclairage (CIE) color coordinates.

Table 2.

TBW2, ABW and CIE coordinates of different ratios of blue emission in white light.

$\Phi_{B0}/(\Phi_{B0} + \Phi_{Y0})$	TBW2/MHz	ABW/MHz	CIE (x, y)
29.8%	9.8	9.5	(0.372, 0.386)
42.3%	10.1	9.8	(0.323, 0.341)
58.2%	10.5	10.3	(0.271, 0.270)