Unique sandwich design of high-efficiency heat-conducting phosphor-in-glass film for high-quality laser-driven white lighting

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Abstract: Multi-color phosphor-in-glass (PiG) film has been considered as a promising color converter in high-quality laser lighting owing to its outstanding merits of phosphor versatility, tunable luminescence, and simple preparation. However, the opto-thermal properties of PiG film are severely affected by the photon reabsorption and backward scattering of phosphor structure and the heat conduction of substrate. Herein, a unique sandwich design of phosphor structure was introduced in the multi-color PiG film for high-quality laser lighting. By elaborately synthesizing the borosilicate glass with low glass transition temperature (Tg), similar expansion coefficient, and high refractive index (RI), the sandwiched PiGs were prepared by sintering (~600 ℃) broadband green and red phosphor glass films on the double sides of sapphire. The green and red PiG films were tightly coated on the sapphire with no delamination and maintained higher luminescence intensity than raw phosphors at high temperatures. By simultaneously coupling photon reabsorption and backward scattering, the sandwiched green PiG film–sapphire–red PiG film (G–S–R PiG) yields a high-quality white light with a high luminous efficacy of 163 lm/W and an excellent color rendering index (CRI) of 85.4 under a laser power of 2.4 W, which are the best comprehensive results yet reported. Benefiting from the ingenious sandwich design with heat-conducting sapphire and thin PiG films, the G–S–R PiG displays low working temperatures (< 200 ℃) under high-power laser excitation. This work reveals the role of sandwiched phosphor structure in photon loss and heat dissipation, which provides a new strategy to design PiG films for high-quality laser lighting.

Keywords: laser-driven lighting; phosphor-in-glass (PiG) film; sandwich structure; color quality; heat dissipation

1 Introduction

Laser-driven white lighting has been treated as a next-generation high-brightness solid-state light source in automotive headlights, laser television (TV), projectors, medical health, and visible light communication [1–4]. Generally, white laser diode (LD) can be produced by a laser-driven phosphor technology, in which a remote color converter is excited by a blue LD [5–8]. Compared with the blue light-emitting diode (LED), the blue LD
has unique characteristics of no efficiency drop, low beam divergence, and high output power density [9,10]. In the meanwhile, it brings serious difficulty to the design and preparation of color converter for the suffering of high laser radiation energy. Due to its poor heat resistance and low thermal conductivity, the traditional color converter of phosphor-in-resin (PiS) has some heat-induced problems of erosion and carbonization under high-power-density laser excitation, which leads to the thermal failure of white LDs [11–15].

To solve the thermal failure from phosphor materials, three types of inorganic color converters have been developed to replace the traditional PiS converter, including single crystals (SCs) [16,17], transparent ceramics (TCs) [18–20], and phosphor-in-glasses (PiGs) [21–23]. These inorganic color converters have been recognized as effective phosphor materials in laser-driven lighting due to their high heat resistance and excellent luminescence. Although the SCs and TCs have high thermal conductivity, they require complicated and costly preparation process [24,25]. It is difficult to recover red and cyan emission spectra originating from the limited base materials of luminescence center in the SCs and TCs, which deteriorates the color quality of white LDs [26,27]. On the contrary, the PiGs avoid these drawbacks owing to the competitive advantages of simple preparation, low cost, phosphor versatility, and tunable luminescence [28–30]. It should be noted that the low thermal conductivity of glass matrix gives rise to the easy luminescence saturation for the PiG converter. At present, another type of PiG has been prepared by sintering a phosphor-glass film on a transparent or reflected substrate, such as glass, sapphire, aluminum, and ceramic substrates [31–33]. The PiG film still maintains the outstanding merits of PiG; meanwhile, it displays high luminescence saturation owing to the efficient heat conduction of thick film and the high thermal conductivity of substrate.

Initially, single yellow-emitting or green-emitting phosphor (e.g., Y₃Al₅O₁₂:Ce³⁺ (YAG), La₃Si₆N₁₃:Ce³⁺ (LSN), or Lu₂Al₅O₁₂:Ce³⁺ (LuAG)) has been used in laser-driven white lighting [34–37]. The PiG film with single phosphor limits the color quality of white LDs due to the lack of red and cyan spectra, resulting in low color rendering index (CRI) (< 65) and high correlated color temperature (CCT) (> 9000 K). Aiming to this issue, some red-emitting phosphors have been added to compensate the red spectrum in the PiG film, such as Ca₅AlSi₃N₅:Eu²⁺ (CASN) and K₅Si₃F₁₃:Mn⁴⁺ (KSFM) phosphors [38–40]. In addition, a green-emitting Y₃Al₃.0₈Ga₁.₉₂O₁₂:Ce³⁺ (YAGG) phosphor with a broad emission has been used in the PiG film for the compensation of cyan spectrum [41,42]. Although the multi-color PiG films can enhance the color rendering of white LDs, the red-emitting phosphor has severe photon reabsorption in green and yellow emission region, which reduces the luminous efficacy of white LDs [43,44]. In Refs. [45,46], the multilayer and pattern structure design of PiG film has been developed to eliminate the photon reabsorption between phosphors. The multilayer PiG film is prepared by stacking a yellow or green phosphor layer on a red phosphor layer, which yields higher luminescence than the mixed PiG film [47,48]. Notably, the photon reabsorption must be considered with the luminous efficacy of radiation (LER) of dominant phosphor layer in the multilayer structure [49]. On the other hand, the PiG film is designed in the pattern structure with a sector piece or concentric ring phosphor geometry, which reduces the spectral overlap of phosphors, and then enhances luminous efficacy and color uniformity. For example, Meng et al. [50] prepared a multilayer PiG film with green-emitting and orange-emitting PiG film on the two sides of sapphire, which exhibits high luminous emittance and better heat-dissipation in laser lighting. Huang et al. [51,52] prepared a PiG film with pattern structure to inhibit photon reabsorption, and this converter further served as a motor-driven rotatory “phosphor wheel” to reduce thermal load under high-power blue LD driven. Unfortunately, the PiG film has serious backward scattering light (higher than 50% of emission light) in the transmissive white LDs, which not only affects the photon reabsorption of phosphors, but also reduces the luminous flux and efficiency of actual forward illumination [53–55]. Ma and Luo [56] developed the secondary optics consisting of a parabolic reflector coupled with a freeform lens, which enhances the luminance of white LDs by nearly four times owing to the collection of backward scattering light and the reduction of divergent angle. Zheng et al. [57] developed a PiG film–sapphire coated with one-dimensional photonic crystals (1DPCs), which decreases the reflection loss of incident blue laser light and reflects the backward emission from phosphors. It should be noted that the opto-thermal properties of PiG film are greatly influenced by the backward blue light generated from the scattering of phosphors and pores and the interface reliability of PiG film and sapphire. However,
the photon reabsorption and backward scattering of phosphor structure has not been studied simultaneously in laser-driven white lighting so far. Importantly, the multi-color PiG film displays low heat dissipation due to the increase thickness and residual pores of film [58]. It is of great interest to design and develop a heat-conducting PiG film with efficient phosphor structure for high-quality laser-driven white lighting.

In this work, we proposed a unique sandwich design of high-efficiency heat-conducting PiG film for high-quality laser-driven white lighting. The photon reabsorption and backward scattering of sandwiched PiGs were studied in detail. The sandwiched green PiG film–sapphire–red PiG film (G–S–R PiG) was prepared by sintering broadband green-emitting YAGG and red-emitting CASN PiG films on the double sides of sapphire substrate. The sapphire was treated as the intermediate for this transmissive PiG owing to its high thermal conductivity (~30 W/(m·K)), high transmittance (~90%), and superior mechanical property [59,60]. The microstructures and luminescence of sandwiched PiGs were characterized. The sandwiched PiGs with different G-PiG film thicknesses were excited by a high-power blue LD, and their optical performance was evaluated. The thermal performance of G–S–R PiG under high-power laser excitation was compared with those of the traditional PiGs.

2 Experimental

2.1 Materials

Commercial green-emitting YAGG phosphor and red-emitting CASN phosphor (Grirem Co. Ltd., China) were treated as the raw materials. The YAGG phosphor particles display an elliptical contour with a mean grain size of 16 μm, and the CASN phosphor particles have a strip shape with a mean grain size of 20 μm (Fig. S1 in the Electronic Supplementary Material (ESM)). The borosilicate glass powders with a chemical composition of 25B2O3–10SiO2–35ZnO–6Li2O–12La2O3–12WO3 (in mol%) were synthesized by the melting–quenching method, as detailed in Ref. [61]. The polished single-crystal sapphires (Zhaohong Technol. Co., Ltd., China) with a thickness of 0.5 mm (the circular structure with a diameter of 18 mm and the square structure with a length of 18 mm) were used as the heat-conducting substrate. The organic solvent was prepared by sufficiently dissolving ethyl cellulose in terpilenol (Aladdin, China) at 80 °C for 5 h.

2.2 Fabrication of sandwiched G–S–R PiG converter

Figure 1 shows the preparation process of sandwiched PiG film converter for laser-driven white lighting. First, a phosphor glass paste was obtained by mixing phosphor particles, glass powders, and organic solvent. The mass ratios of YAGG phosphor to glass and CASN phosphor to glass were 50 and 10 wt%, respectively. Then, the green phosphor glass paste was coated on one surface of sapphire and dried at 120 °C for 15 min. After that, the red phosphor glass paste was coated on the other surface of sapphire and dried at 120 °C for 15 min. The thickness of paste coating was precisely controlled by varying the space between scraper and sapphire. Finally, the sandwiched G–S–R PiG was achieved after the paste-coated sapphire sintered at 600 °C for 0.5 h and annealed at 300 °C for 1 h. The thickness of G-PiG film was adjusted to tune the luminescence of G–S–R PiG. The green PiG film–red PiG film–sapphire (G–R–
S PiG) and mixed green/red PiG film–sapphire (M–S PiG) were prepared by the same mass ratio of phosphors, which were treated as the comparison samples. In addition, the white LDs were fabricated by combining the sandwiched PiG converters and blue LDs (λ = 445 nm). The blue laser light was collimated and focused on the PiG by a lens system, and the blue light and phosphor-converted green/red light were mixed to realize white emission light. The secondary optics with a parabolic reflector was integrated in the sandwiched PiG to collect the backward scattering light, resulting in sufficient forward emission light for illumination.

2. 3 Characterizations

The microstructures and element distributions of G–S–R PiGs and their phosphors were observed by a scanning electron microscope (SEM; Nova NanoSEM 450, FEI, USA) combined with an energy dispersive spectrometer. The X-ray diffraction (XRD) patterns of G-PiG, R-PiG, and related phosphors were determined by an X-ray diffractometer (X’Pert PRO MRD, PANalytical, the Netherlands) equipped with a Cu-Kα radiation source in a range of 2θ = 15°–80°. The photoluminescence excitation (PLE), PL spectra, fluorescence lifetime, and temperature-dependent PL spectra (25–205 °C) of G-PiG, R-PiG, and related phosphors were measured by a fluorescence spectrometer (FLS1000, Edinburgh Instruments, UK); the holding time at each temperature point was set as 4 min, and the heating rate was controlled as 50 °C/min. The transmission spectra of G–S–R PiGs were characterized, as presented in Fig. 2. The G-PiG and R-PiG films are tightly coated on two sides of sapphire, and no obvious delamination can be observed in the film–sapphire interfaces. The G-PiG and R-PiG films achieve the same thickness of about 70 μm. It can be seen that the YAGG and CASN phosphor particles are well embedded in the glass matrix (Fig. S4 in the ESM). The R-PiG film displays a smooth surface without pores, while the G-PiG film has some pores from surface to inside, which is attributed to more particle interval from the high mass ratio of green phosphor in the G-PiG film. The appropriate scattering of pores is prone to promoting the absorption and conversion of phosphor, and improving the color uniformity of emission light. The in-line energy dispersive spectroscopy (EDS) spectra on the embedded YAGG and CASN phosphor particles display intense signals of Y element and Ca element from their chemical component, while the signal of glass component (Zn element) is very weak. The SEM–EDS mappings present that the Ca, Al, Si, and N elements were detected on the G-PiG film. The appropriate scattering of pores is prone to promoting the absorption and conversion of phosphor, and improving the color uniformity of emission light.

The XRD patterns of G-PiG and R-PiG display that some diffraction peaks are almost identical to their phosphors, revealing no obvious difference from those of the standard patterns, as shown in Fig. 3(a). These PiGs have no impurity phase except the amorphous peak from glass matrix, indicating that the phosphor crystals maintain intact structure in the glass matrix. The G-PiG has an excitation peak centered at about 462 nm, while the R-PiG exhibits an excitation peak centered at approximately 467 nm (Fig. 3(b)), which is derived from the 4f → 5d electron transitions of Ce3+ and Eu2+ ions in the YAGG and CASN phosphors, respectively. The G-PiG and R-PiG display the typical Ce3+ and Eu2+ ions’ 5d → 4f broadband emission centered at about 516 and 657 nm under the blue light
(450 nm) excitation, respectively. Importantly, the G-PiG achieves a high full-width at half maximum (FWHM) of 90 nm, which compensates for a cyan spectrum of emission light and contributes to a high color rendering for white light. It is noted that the PLE and PL spectra of G-PiG and R-PiG are similar to those of raw phosphors, indicating that the prepared PiGs maintain the luminescent properties of phosphors. In Figs. 3(c) and 3(d), the decay curves exhibit that the YAGG phosphor has a negligible lifetime change from 56.54 to 55.25 ns, and the CASN phosphor has a lifetime drop from 282.62 to 215.76 ns before and after the PiG preparation process, respectively. The G-PiG exhibits a short decay time (~55 ns), resulting in the low optical excitation quenching of YAGG phosphor under high-power-density laser excitation. Meanwhile, the external quantum efficiency (EQE) of YAGG phosphor decreases from 96% to 90%, while that of CASN phosphor reduces from 89% to 70%. The reduction of quantum efficiency from the phosphor to the PiG is caused by the thermal quenching, in which the energy of the excited state gets back to the ground state through non-radiative transition [62,63]. The high QE reduction of CASN phosphor can be explained by the serious effect of thermal degradation owing to the possible chemical decomposition of nitride phosphor and the oxidation of Eu$^{2+}$ into Eu$^{3+}$ during the sintering process [64]. Considering the importance of the thermal quenching of laser-driven phosphors, the temperature-dependent PL spectra of PiGs and phosphors are measured, as presented in Figs. 3(e)–3(g). When the temperature increases from 25 to 205 °C, the emission intensities of PiGs and phosphors are gradually reduced originating from the non-radiative transition induced by high temperatures [65]. Notably, the G-PiG and R-PiG display higher thermal stability than the related phosphors, which is attributed to the heat resistance of glass matrix and the heat conduction of sapphire. The luminescence intensities of G-PiG and R-PiG still maintain 82.3% and 76.1%, respectively, at 205 °C, while those of green and red phosphors are only 63.8% and 66%, respectively.
3.2 Optical performance of sandwiched PiGs excited by blue LD

The sandwiched G–S–R PiGs with different G-PiG film thicknesses were pumped by the high-power blue LD, and their optical performance was evaluated. The transparency of G–S–R PiGs is gradually reduced with the increase of G-PiG film thickness owing to the enlarged scattering effect of phosphor and pores, as shown in Fig. 4(a). When the thickness of G-PiG film
increases from 40 to 90 μm, the green emission intensity gradually increases, and the blue emission intensity decreases, as presented in Fig. 4(b), ascribing to that more incident blue laser is absorbed by the YAGG phosphor and converted the green emission light. On that account, the luminous flux and luminous efficacy of G–S–R PiGs increase linearly from 300.9 to 513.2 lm and 96 to 163 lm/W under a laser power of 2.4 W, respectively. The corresponding color quality of emission light has some variations, in which the CCT decreases to 6405 K, the CRI changes from 68.6 to 85.4, and the CIE chromaticity coordinates vary from blue–white region to green–white region, as displayed in Figs. 4(c)–4(e). The surface color of G–S–R PiGs changes from orange to yellow, and their transparency gradually degrades (the inset of Fig. 4(e)), which stays the same with the measured transmission spectra. Consequently, the G–S–R PiG with a G-PiG film thickness of 70 μm enables a high-quality white light with a high CRI of 84.3, an appropriate CCT of 7947 K, and a perfect chromaticity coordinate of (0.2945, 0.3092).

In order to evaluate the photon reabsorption and backward scattering of multi-color phosphor structure, the optical performance of G–S–R PiGs with a fixed G-PiG film thickness of 70 μm was studied under various laser powers. In the sandwiched PiG-based white LDs, the incident blue laser was forward focused on the G-PiG films integrated with/without the parabolic reflector, which are treated as the G–S–R PiG and G–S–R-1 PiG samples, respectively. On the contrary, the blue laser focused on the R-PiG films with/without the parabolic reflector are defined as the R–S–G PiG and R–S–G-1 PiG samples, respectively. The G–S–R PiG exhibits higher green emission intensity, while the R–S–G PiG has higher red emission intensity (Fig. 5(a)), attributing to that more blue laser light is absorbed by the close G-PiG and R-PiG films, and then converted to green and red lights, respectively. Meanwhile, the G–S–R and R–S–G PiGs achieve higher green and red emission intensities than the G–S–R-1 and R–S–G-1 PiGs owing to the collection of backward scattering light by the parabolic reflector. As the laser power increases from 0.3 to 5.6 W, the luminous fluxes and CRIs of the four samples are shown in Figs. 5(b) and 5(c), respectively. The luminous fluxes of the G–S–R and R–S–G PiGs are rapidly raised from 21.2 to 786.4 lm and 11.8 to 299.6 lm, respectively; while those of G–S–R-1 and R–S–G-1 PiGs are slowly increased from 0.2 to 171 lm and 0.15 to 149 lm, respectively. The corresponding CRIs of G–S–R and G–S–R-1 PiGs maintain at about 84 and 79, respectively; while the R–S–G and R–S–G-1 PiGs yield an abominable color quality with the CRI lower than 60. It can be seen that a cool-white light emits from the forward and backward of G–S–R PiG, while a cool-white light and a blue–white light emit from the forward and backward of R–S–G PiG, respectively, as presented in Fig. 5(d). Deeply, the light transmission paths in the G–S–R and R–S–G PiGs are explained in Fig. 5(e). For the sandwiched PiGs, a great deal of blue
The laser is absorbed and scattered by the first PiG film, which is close to the blue LD excitation. The luminescence of sandwiched PiGs is mainly determined by the properties of the first PiG film, such as the LER of phosphors, the phosphor content, and the porosity of PiG film. It should be noted that the sandwiched PiGs exhibit forward and backward emission light under high-power laser excitation, and the backward light is higher than the forward light owing to the severe scattering effect of phosphor particles and pores. On this account, the G–S–R PiG has the reabsorption of forward green light by the red phosphor, while the R–S–G PiG has the reabsorption of backward green light by the red phosphor. Although the reabsorption loss reduces the luminescence, the G-PiG film yields higher LER than the R-PiG film, which leads to a higher luminous flux for the G–S–R PiG. Furthermore, the luminous flux of G–S–R PiG is obviously higher than that of G–S–R–1 PiG (565.5 and 126.8 lm at 4.1 W, respectively, nearly 4.5 times), while the luminous flux difference of R–S–G PiG and R–S–G–1 PiG is not huge (nearly 2 times at 4.1 W). It is ascribed to that a lot of incident blue light and phosphor-converted light are backscattered by the phosphors and pores. The G-PiG film with high phosphor contents and porosities leads to more blue and green backward light, which reduces the forward luminescence of G–S–R–1 PiG.

Since the backward light is collected to enlarge forward emission by the parabolic reflector, the G–S–R PiG achieves the highest luminous flux and luminous efficacy in these PiG converters.

On the other hand, the optical performance and actual illumination of G–S–R PiG were compared with those of the M–S and G–R–S PiGs under the blue LD with a laser power of 2.4 W. The G–S–R and G–R–S PiG exhibit higher green intensities and lower red intensities than the M–S PiG as presented in Fig. 6(a). It is attributed to that the red phosphor in the M–S PiG directly absorbs the blue laser and reabsorbs a part of green light, resulting in the increase of red emission and decrease of green emission. The G–S–R PiG has higher green intensity and lower red intensity compared with the G–R–S PiG. The G-PiG film is close to the R-PiG film in the G–R–S PiG, which causes that the green light from the G-PiG film is easily reabsorbed by the R-PiG film and converts to the red light. Correspondingly, the green light of G–S–R PiG has the reflection effect in two interfaces of sapphire, which reduces the reabsorption probability of green light. In Figs. 6(b)–6(d), the G–S–R PiG enables a high luminous efficacy of 141 lm/W, which is enhanced by 77% and 41% compared with the M–S and G–R–S PiGs, respectively. The M–S and G–R–S PiGs achieve low CCTs of 3562 and 5233, respectively, owing to their

![Fig. 6](image-url) (a) EL spectra, (b) luminous fluxes and luminous efficacy, (c) CCTs and CRIs, and (d) CIE chromaticity coordinates of M–S, G–R–S, and G–S–R PiGs under the blue LD with a laser power of 2.4 W. (e) Demonstration experiment of illuminating toy blocks under the M–S PiG-, G–R–S PiG-, and G–S–R PiG-based white LD sources. The thicknesses of G-PiG, R-PiG, and M-PiG films maintain the same thickness of 70 μm.
high red emission intensity. The CRI of G–S–R (~85) PiG is higher than those of M–S (~75) and G–R–S (~80) PiGs. The G–S–R PiG exhibits a perfect chromaticity coordinate located on the Planckian locus, while the chromaticity coordinates of M–S and G–R–S PiGs are away from the Planckian locus. In Fig. 6(e), the toy blocks with various color bodies are illuminated by the M–S PiG-, G–R–S PiG-, and G–S–R PiG-based white LD sources. The G–S–R PiG enables high color quality for white LD source, which reproduces the toy blocks with more vivid and bright color.

3.3 Thermal performance of sandwiched PiGs excited by blue LD

The thermal performance of G–S–R PiG under high-power laser excitation was evaluated and compared with those of the M–S and G–R–S PiGs. As the laser power increases from 0.3 to 4.1 W, the maximum surface temperature of G–S–R PiG slowly rises from 29 to 129 °C, while those of M–S and G–R–S PiGs increase from 30 to 157 °C and 34 to 289 °C, respectively, as displayed in Fig. 7(a). It is attributed to that the G–S–R PiG realizes the simultaneous heat conduction of both-sided PiG films, and the M–S PiG has a thin PiG film contributed to heat dissipation compared with the G–R–S PiG. The surface temperatures of three PiGs rapidly rise before the laser driven time of 40 s and tend to be stable after 120 s. At a laser power of 2.4 W, the maximum surface temperatures of M–S, G–R–S, and G–S–R PiGs stabilize at 104, 156, and 93 °C after the laser driven time of 120 s, respectively (Fig. 7(b)). In Figs. 7(c) and 7(d), the G–S–R PiG yields stable luminous flux (loss of ~1%) under the blue LD with a laser power of 4.1 W after 300 s, while the luminous flux losses of M–S and G–R–S PiGs reach up to 5% and 26%, respectively. The G–R–S PiG exhibits an obvious reduction of CRI from 80.5 to 75.5. The luminescence degradation of G–R–S PiG is ascribed to the thermal quenching of phosphors from a high working temperature of 289 °C, as presented in Fig. 7(e). In view of this, the ingenious sandwich design of phosphor structure enhances the heat dissipation of PiGs under high-power laser excitation owing to the heat-conducting sapphire between two-sided PiG films and the thin thickness of PiG films.

4 Conclusions

In this work, the sandwiched PiG converters were prepared for high-quality laser lighting. The borosilicate glass matrix was carefully synthesized with low $T_g$,
similar expansion coefficient, and high RI. The broadband green and red PiG films were tightly sintered on the double sides of sapphire, which has no delamination in the film–sapphire interfaces. The green and red PiG films display higher thermal stability than the raw phosphors, and still maintain remarkable luminescence intensity at high temperatures. The G-PiG film thickness of G–S–R PiGs was adjusted to optimize the optical performance of white LDs. When the thickness of G-PiG film increases from 40 to 90 μm, the luminous flux and luminous efficacy of G–S–R PiGs increase linearly from 300.9 to 513.2 lm and 96 to 163 lm/W under a laser power of 2.4 W, respectively. By simultaneously coupling photon reabsorption and backward scattering, the G–S–R PiG exhibits higher luminous flux and luminous efficacy compared with the M–S and G–R–S PiGs. The G–S–R PiG enables a high-quality white light with a high CRI of 84.3, an appropriate CCT of 7947 K, and a perfect chromaticity coordinate of (0.2945, 0.3092). Remarkably, the G–S–R PiG exhibits higher luminous flux and luminous efficacy compared with the M–S and G–R–S PiGs. The G–S–R PiG enables a high-quality white light with a high CRI of 84.3, an appropriate CCT of 7947 K, and a perfect chromaticity coordinate of (0.2945, 0.3092). Remarkably, the G–S–R PiG has low working temperatures (< 200 °C) under high-power laser excitation ascribing to the double-sided heat-conduction of sapphire and the thin thickness of PiG films. The results indicate that the sandwiched PiG film is the high-efficiency heat-conducting color converter for high-brightness and high-quality laser-driven white lighting.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material

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References

[1] Tsao JY, Crawford MH, Coltrin ME, et al. Toward smart and ultra-efficient solid-state lighting. Adv Opt Mater 2014, 2: 809–836.
[2] Chi YC, Hsieh DH, Lin CY, et al. Phosphorous diffuser diverged blue laser diode for indoor lighting and communication. Sci Rep 2015, 5: 18690.
[3] Schütt F, Zapf M, Signetti S, et al. Conversionless efficient and broadband laser light diffusers for high brightness illumination applications. Nat Commun 2020, 11: 1437.
[4] Song YH, Kwon SB, Jung MK, et al. Fabrication design for a high-quality laser diode-based ceramic converter for a laser headlamp application. Ceram Int 2018, 44: 1182–1186.
[5] Krasnoshchoka A, Hansen AK, Thorseth A, et al. Phosphor material dependent spot size limitations in laser lighting. Opt Express 2020, 28: 5758–5767.
[6] Ma YP, Luo XB. Packaging for laser-based white lighting: Status and perspectives. J Electron Packaging 2020, 142: 010801.
[7] Park J, Kim J, Kwon H. Phosphor–aluminum composite for energy recycling with high-power white lighting. Adv Opt Mater 2017, 5: 1700347.
[8] Yeh CT, Chou YI, Yang KS, et al. Luminescence material characterizations on laser-phosphor lighting techniques. Opt Express 2019, 27: 7226–7236.
[9] Wierer JJ, Tsao JY, Sizov DS. Comparison between blue lasers and light-emitting diodes for future solid-state lighting. Laser Photonics Rev 2016, 7: 963–993.
[10] Wierer JJ, Tsao JY. Advantages of III-nitride laser diodes in solid-state lighting. Phys Status Solidi A 2015, 212: 980–985.
[11] Xie B, Hu R, Luo XB. Manipulating heat transport of photoluminescent composites in LEDs/LDs. J Appl Phys 2021, 130: 070906.
[12] Ding XR, Li M, Li ZT, et al. Thermal and optical investigations of a laser-driven phosphor converter coated on a heat pipe. Appl Therm Eng 2019, 148: 1099–1106.
[13] Yan C, Ding X, Chen M, et al. Research on laser illumination based on phosphor in metal (PiM) by utilizing the boron nitride-coated copper foams. ACS Appl Mater Interfaces 2021, 13: 29996–30007.
[14] Lin SS, Lin H, Chen GX, et al. Stable CsPbBr2−xI2x-glass nanocomposite for low-étendue wide-color-gamut laser-driven projection display. Laser Photonics Rev 2021, 15: 2100044.
[15] Wang PF, Sui P, Lin SS, et al. Lu2SrAl5SiO12 :Ce3+ phosphor in glass film-on-sapphire and its application to laser lighting. Chin J Lumin 2021, 42: 1493–1501. (in Chinese)
[16] Park KW, Lim SG, Deressa G, et al. High power and temperature luminescence of Y2Al2O5 :Ce3+ bulky and pulverized single crystal phosphors by a floating-zone method. J Lumin 2015, 168: 334–338.
[17] Cantore M, Pfaff N, Farrell RM, et al. High luminous flux
from single crystal phosphor-converted laser-based white lighting system. Opt Express 2016, 24: A215–A221.

[18] Yao Q, Hu P, Sun P, et al. YAG:Ce³⁺ transparent ceramic phosphors brighten the next-generation laser-driven lighting. Adv Mater 2020, 32: 1907888.

[19] Zheng P, Li SX, Wei R, et al. Unique design strategy for laser-driven color converters enabling superhigh-luminance and high-directionality white light. Laser Photonics Rev 2019, 13: 1900147.

[20] Li YB, Ma CY, Zuo CD, et al. High thermal stability AlN–YAG:Ce composite phosphor ceramics for high-power laser-driven lighting. Appl Phys Lett 2021, 119: 251903.

[21] Peng Y, Mou Y, Wang H, et al. Stable and efficient all-inorganic color converter based on phosphor in tellurite glass for next-generation laser-excited white lighting. J Eur Ceram Soc 2020, 38: 5525–5532.

[22] Lin T, Chen HX, Li SX, et al. Bi-color phosphor-in-glass films achieve superior color quality laser-driven stage spotlights. Chem Eng J 2022, 444: 136591.

[23] Mou Y, Yu ZK, Lei ZY, et al. Enhancing opto-thermal performances of white laser lighting by high reflective phosphor converter. J Alloys Compd 2022, 918: 165637.

[24] Chen DQ, Xiang WD, Liang XJ, et al. Advances in transparent glass-ceramic phosphors for white light-emitting diodes—A review. J Eur Ceram Soc 2015, 35: 859–869.

[25] Li SX, Wang L, Hirosaki N, et al. Color conversion materials for high-brightness laser-driven solid-state lighting. Laser Photonics Rev 2018, 12: 1800173.

[26] Peng Y, Mou Y, Zhuo Y, et al. Preparation and luminescent performances of thermally stable red-emitting phosphor-in-glass for high-power white lighting. J Alloys Compd 2018, 768: 114–121.

[27] Zhang YJ, Liang YY, Zhang YQ, et al. High color rendering index composite phosphor-in-glass for high-power white laser lighting. J Eur Ceram Soc 2021, 41: 4915–4923.

[28] Lin H, Hu T, Cheng Y, et al. Glass ceramic phosphors: Towards long lifetime high-power white light-emitting-diode applications—A review. Laser Photonics Rev 2018, 12: 1700344.

[29] Zhang D, Xiao W, Liu C, et al. Highly efficient phosphor-glass composites by pressureless sintering. Nat Commun 2020, 11: 2805.

[30] Ma CY, Cao YG. Phosphor converters for laser driven light sources. Appl Phys Lett 2021, 118: 210503.

[31] Xu J, Liu BG, Liu ZW, et al. Design of laser-driven SiO₂–YAG:Ce composite thick film: Facile synthesis, robust thermal performance, and application in solid-state laser lighting. Opt Mater 2018, 75: 508–512.

[32] Wang H, Mou Y, Peng Y, et al. Fabrication of phosphor glass film on aluminum plate by using lead-free tellurite glass for laser-driven white lighting. J Alloys Compd 2020, 814: 152321.

[33] Liu ZH, Hu P, Jiang HJ, et al. CaAlSiN3:Eu³⁺/Lu₃AlO₂:Ce³⁺ phosphor-in-glass film with high luminous efficiency and CRI for laser diode lighting. J Mater Chem C 2021, 9: 3522–3530.

[34] Zhang XJ, Si SC, Yu JB, et al. Improving the luminous efficacy and resistance to blue laser irradiation of phosphor-in-glass based solid state laser lighting through employing dual-functional sapphire plate. J Mater Chem C 2019, 7: 354–361.

[35] You SH, Li SX, Zhang P, et al. A thermally robust La₃Si₆N₁₄:Ce-in-glass film for high-brightness blue-laser-driven solid state lighting. Laser Photonics Rev 2019, 13: 1800216.

[36] Yang HS, Zhang YJ, Zhang YQ, et al. Designed glass frames full color in white light-emitting diodes and laser diodes lighting. Chem Eng J 2021, 414: 128754.

[37] Yue XM, Xu J, Lin H, et al. β-Sialon:Eu³⁺ phosphor-in-glass film: An efficient laser-driven color converter for high-brightness white color-gamut projection displays. Laser Photonics Rev 2021, 15: 2100317.

[38] Zhang XJ, Yu JB, Wang J, et al. All-inorganic light converter based on phosphor-in-glass engineering for next-generation modular high-brightness white LEDs/LDs. ACS Photonics 2017, 4: 986–995.

[39] Zhu QQ, Xu X, Wang L, et al. A robust red-emitting phosphor-in-glass (PiG) for use in white lighting sources pumped by blue laser diodes. J Alloys Compd 2017, 702: 193–198.

[40] Si SC, Huang L, Zhang XJ, et al. A stable and efficient red-emitting color converter based on K₂SiF₆:Mn⁴⁺ phosphor-in-glass film for next-generation high-power WLEDs. ACS Appl Electron Mater 2020, 2: 2929–2936.

[41] Peng Y, Wang H, Liu JX, et al. Broad-band and stable phosphor-in-glass enabling ultrahigh color rendering for all-inorganic high-power WLEDs. ACS Appl Electron Mater 2021, 3: 2100146.

[42] Zhang JH, Wang LH, Zhu QQ, et al. A thermally robust phosphor-in-glass film with high luminous efficiency for high-power blue laser diodes lighting. Appl Phys Lett 2021, 119: 221904.

[43] Chiang CH, Tsai HY, Zhan TS, et al. Effects of phosphor distribution and step-index remote configuration on the performance of white light-emitting diodes. Opt Lett 2015, 40: 2830–2833.

[44] Zhang YJ, Zhang ZL, Liu XD, et al. A high quantum efficiency CaAlSiN₃:Eu³⁺ phosphor-in-glass with excellent optical performance for white light-emitting diodes and blue laser diodes. Chem Eng J 2020, 401: 125983.

[45] Ying SP, Shen JY. Concentric ring phosphor geometry on the luminous efficiency of white-light-emitting diodes with excellent color rendering property. Opt Lett 2016, 41: 1989–1992.

[46] Peng Y, Li RX, Cheng H, et al. Facile preparation of patterned phosphor-in-glass with excellent luminous properties through screen-printing for high-power white light-emitting diodes. J Alloys Compd 2017, 693: 279–284.

[47] Xiang R, Liang XJ, Li PZ, et al. A thermally stable warm
WLED obtained by screen-printing a red phosphor layer on the LuAG:Ce\(^{3+}\) PiG substrate. *Chem Eng J* 2016, **306**: 858–865.

[48] Peng Y, Sun QL, Liu JX, *et al.* Fabrication of stacked color converter for high-power WLEDs with ultra-high color rendering. *J Alloys Compd* 2021, **850**: 156811.

[49] Lin ZB, Lin H, Xu J, *et al.* Highly thermal-stable warm w-LED based on Ce:YAG PiG stacked with a red phosphor layer. *J Alloys Compd* 2015, **649**: 661–665.

[50] Meng Y, Zhu QQ, Huang MH, *et al.* Sandwich structured phosphor-in-glass films enabling laser lighting with superior optical properties. *Ceram Int* 2022, **48**: 13626–13633.

[51] Huang QG, Sui P, Huang F, *et al.* Toward high-quality laser-driven lightings: Chromaticity-tunable phosphor-in-glass film with “phosphor pattern” design. *Laser Photonics Rev* 2022, **16**: 2200040.

[52] Huang QG, Lin H, Wang B, *et al.* Patterned glass ceramic design for high-brightness high-color-quality laser-driven lightings. *J Adv Ceram* 2022, **11**: 862–873.

[53] Wang L, Wei R, Zheng P, *et al.* Realizing high-brightness and ultra-wide-color-gamut laser-driven backlighting by using laminated phosphor-in-glass (PiG) films. *J Mater Chem C* 2020, **8**: 1746–1754.

[54] Lenef A, Kelso JF, Serre J, *et al.* Co-sintered ceramic converter for transmissive laser-activated remote phosphor conversion. *Appl Phys Lett* 2022, **120**: 021104.

[55] Cao YX, Chen W, Du YJ, *et al.* Luminous performances characterization of YAG:Ce\(^{3+}\) phosphor/silicone composites using both reflective and transmissive laser excitations. *IEEE Photonics J* 2022, **14**: 8216706.

[56] Ma YP, Luo XB. Small-divergent-angle uniform illumination with enhanced luminance of transmissive phosphor-converted white laser diode by secondary optics design. *Opt Laser Eng* 2019, **122**: 14–22.

[57] Zheng P, Li SX, Wang L, *et al.* Unique color converter architecture enabling phosphor-in-glass (PiG) films suitable for high-power and high-lumiance laser-driven white lighting. *ACS Appl Mater Interfaces* 2018, **10**: 14930–14940.

[58] Wei R, Wang L, Zheng P, *et al.* On the luminescence saturation of phosphor-in-glass (PiG) films for blue-laser-driven white lighting: Effects of the phosphor content and the film thickness. *J Eur Ceram Soc* 2019, **39**: 1909–1917.

[59] Peng Y, Mou Y, Sun QL, *et al.* Facile fabrication of heat-conducting phosphor-in-glass with dual-sapphire plates for laser-driven white lighting. *J Alloys Compd* 2019, **790**: 744–749.

[60] Zhang XJ, Si SC, Yu JB, *et al.* Improving the luminous efficiency and resistance to blue laser irradiation of phosphor-in-glass based solid state laser lighting through employing dual-functional sapphire plate. *J Mater Chem C* 2019, **7**: 354–361.

[61] Mou Y, Wang H, Liang DD, *et al.* Efficient and heat-conducting color converter of phosphor glass film printed on sapphire substrate for high-power white LEDs/LDs. *J Non-Cryst Solids* 2019, **515**: 98–105.

[62] Yue XM, Lin H, Lin SS, *et al.* La\(_3\)Si\(_6\)N\(_{11}\):Ce\(^{3+}\) luminescent glass ceramics applicable to high-power solid-state lighting. *Chin J Lumin* 2020, **41**: 1529–1537. (in Chinese)

[63] Zhang YJ, Liang YY, Zhang YQ, *et al.* High color rendering index composite phosphor-in-glass for high-power white laser lighting. *J Eur Ceram Soc* 2021, **41**: 4915–4923.

[64] Zhang JD, Wang LS, Xu FC, *et al.* High-efficiency phosphor-in-glass with ultra-high color rendering indexing for white laser diode lighting. *Ceram Int* 2022, **48**: 1682–1689.

[65] Zheng P, Li SX, Takeda T, *et al.* Unraveling the luminescence quenching of phosphors under high-power-density excitation. *Acta Mater* 2021, **209**: 116813.

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