Journal of Luminescence 181 (2017) 184–188

Contents lists available at ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

Full length article

Emission enhancements in phosphor chips with surface gratings

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ARTICLE INFO

Article history:
Received 9 November 2015
Received in revised form
29 August 2016
Accepted 5 September 2016
Available online 12 September 2016

Keywords:
Solid state lighting
Surface coating
Extraction efficiency

ABSTRACT

An oxide grating fabricated with standard microlithographic processes and patterned with interference lithography was fabricated on a solid LuAG:Ce^{3+} phosphor chip about 0.2 mm thick. Integrating sphere emission measurement shows an increase of 8.1% in the emission band from 520 to 550 nm, with the majority of the increase coming at steeper emission angles. This process may be a practical way to increase solid state lighting efficiency. This technique was effective even though the phosphor was not optically flat.

1. Introduction

Solid state lighting has made tremendous progress since blue light-emitting diodes were developed in 1994 [1]. Compared with traditional incandescent or fluorescent light, solid state lighting technology is much more efficient and reliable, with lifetimes greater than 10,000 hours and efficiency of >90 lm/W have been realized. It is expected that solid state lighting will eventually be the standard in lighting technology. With the Department of Energy calling for an "efficacy target" of >250 lm/W [2], it is desired to engineer this solid state lighting to be as efficient as possible.

The most common general type of white light bulb consists of a blue light emitting diode (LED) combined with phosphors which luminesce in the yellow or green [3]. The commonly used combination is InGaN blue LED and Ce^{3+} doped $Y_3Al_5O_{12}$ (YAG:Ce^{3+}) [4]. In these systems, the total efficiency depends on both the internal quantum efficiency and the light extraction efficiency.

The usual form factor is phosphor powder encapsulated by silicone, but for higher temperatures and more compact structures, a phosphor platelet directly coupled to the LED is desirable. This solid phosphor is more tolerant of heat than silicone, but the interface between phosphor and air has a greater index difference, which reduces the extraction efficiency of the light out of the phosphor. Light incident at an angle larger than the critical angle of this interface will be totally internally reflected, and so not be coupled out the front of the phosphor. While a good package may eventually recycle many of these scattered photons, it is desirable to enhance the phosphor surface to increase this extraction efficiency.

Recent work has focused on overcoming this extraction limitation, and many different techniques have been applied. Oh et al., achieved a 4*x* improvement in extraction efficiency in a Y_2O_3:Eu+ doped thin film phosphor when coated with a triangular or square photonic crystal pattern thin film of SiN_x [5]. The same technique was later applied to very smooth (roughness ~5 Å) sintered. 0.1 mm thick YAG:Ce3+ film with a 30% enhancement of light extraction [6]. This sort of 2D photonic crystal patterning technique has been shown to increase efficiency in solar cells as well [7]. Self-organized ZnO nanorods were also shown to increase efficiency from GaN-based LEDs coupled to phosphor powders [8]. Coupled nanoparticles have achieved enhanced luminescence by increasing outcoupling to quasi-guided and Rayleigh anomaly modes in similar YAG:Ce substates [9].

Park et al. [10], later reported in a comparison of phosphor form factors (between thick and thin films) an improvement of about 25–30% with a 2D photonic crystal layer. Mao et al., reported a factor of 4.5 improvement in the forward yellow emission with the fabricated TiO_2 photonic crystal layer on a polished YAG:Ce^{3+} substrate [11]. Improvements have been seen in phosphor powder when coating it with silica [12].

These techniques are powerful but have some limitations. So far, results have only been reported on relatively smooth, polished phosphors (5 Å roughness). Both the lithography and the effectiveness may be compromised on substrates with significant roughness.

Prior results have used nanosphere-based lithography or imprint lithography to achieve the <1 μm pitch of the photonic crystals necessary for optimal outcoupling of visible light [13,14].

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http://dx.doi.org/10.1016/j.jlumin.2016.09.018
0022-2313/

Here we describe a technique based on interference lithography that is effective on unpolished phosphor substrates. This is based on significant surface roughening rather than coherent out-scattering. Roughening the surface of the emitting phosphor can improve the light extraction efficiency [15,16]. The large number of facets introduced by roughened surface redirects the internal reflected light back into the escape cone and increases external emission. The surface is roughened by first depositing a layer of SiO_2, which is then patterned into a grating with a ~650 nm pitch. This method achieved a significant wide angle emission enhancement with no observable diffraction effects and was realized with conventional microfabrication tools scalable to large areas.

2. Experiment

The phosphor in this work is $Lu_3Al_5O_{12}:Ce^{3+}$(LuAG:Ce^{3+}), a green phosphor which has better thermal stability and intensity than YAG:Ce^{3+} [17] and whose grain size and morphology are shown in Fig. 1. The phosphor is doped to 1% mol replacement fraction of Lu by Ce. The phosphor chips were produced by forming and sintering methods, and then cutting into the appropriate thickness and shape. For these studies, the chips were 5 mm in diameter and about 0.2 mm thick. The roughness was characterized using atomic force microscopy and was about 100 nm root-mean-square on the 30 μm square sample dimension.

The fabrication flow using a Lloyd's mirror interferometer is outlined in Fig. 2. The LuAG:Ce^{3+} phosphor is very resistant to both chemical and physical etching which makes it difficult to roughen directly. To circumvent this problem, a 100 nm thick SiO_2 layer was grown on the surface using plasma-enhanced chemical vapor deposition. Besides being a convenient material, SiO_2 also has a refractive index of 1.53, and is therefore a good intermediate optical layer between LuAG:Ce^{3+} ($n \sim 1.83$) and air ($n \sim 1$).

After that, a layer of Shipley 1813 photoresist was then spun onto the phosphor. For spinning convenience, the 5 mm diameter phosphor was attached on a 5 in. Si piece, 2 cm away from center, and spun 45 s at 3500 rpm. Then it was exposed in a Lloyd's mirror interferometer. The Lloyd's mirror interferometer used a 425 nm wavelength violet laser with a laser power density of 0.69 mW/cm^2 and an exposure time of 35 s. The stage angle was set to create a grating pattern with a 638 m period.

The resist is then developed with MF-26A developer leaving a grating structure photoresist on the sample (Figs. 3 and 4.). After hardbaking at 100 °C for 60 s, the photoresist served as a mask for reactive ion etching (RIE), to transfer the pattern to the SiO_2 layer. After etching, the surface morphology of LuAG:Ce^{3+} piece was characterized by AFM (atomic force microscopy) (Fig. 3(a)) and SEM (Fig. 4).

To measure the emission of LuAG:Ce^{3+}, the LuAG:Ce^{3+} piece was placed on a fixture with a blue LED emitting at a center wavelength of 450 nm. The fixture had a recess exactly fitting the 5 mm chips enabling the chip to be placed and coupled consistently to the LED. Before each phosphor measurement was taken, a calibration measurement was taken with just the LED at a standard current and integration time, to ensure that the stage was stable and the measurement of output variation was reliable.

Two different sets of measurements were obtained. In the first, the phosphor and LED fixture were placed in the mouth of an integrating sphere from Labsphere Inc. and the emission over all angles collected (Fig. 5(a)). This gives the change in emission regardless of direction. In a second measurement (Fig. 5(b)), the angular distribution of the emission was measured via an optic fiber fixed on a spin stage which could be rotated to collect the light emitted into a given angle. Between the phosphor and the LED, there is a pin hole on top of LED to restrict all the excitation light to couple into the phosphor. In both measurements, a fiber connected to a Jazz Spectroscopy Suite from Ocean Optics was used to collect the light, and the Spectroscopy Suite software used to control the measurement and collect and analyze the data. Angular emission data was measured from 0° to 40° with a 10° degree steps.

Emission from the phosphor was measured before any processing, after deposition of the 100 nm SiO_2, and after patterning.

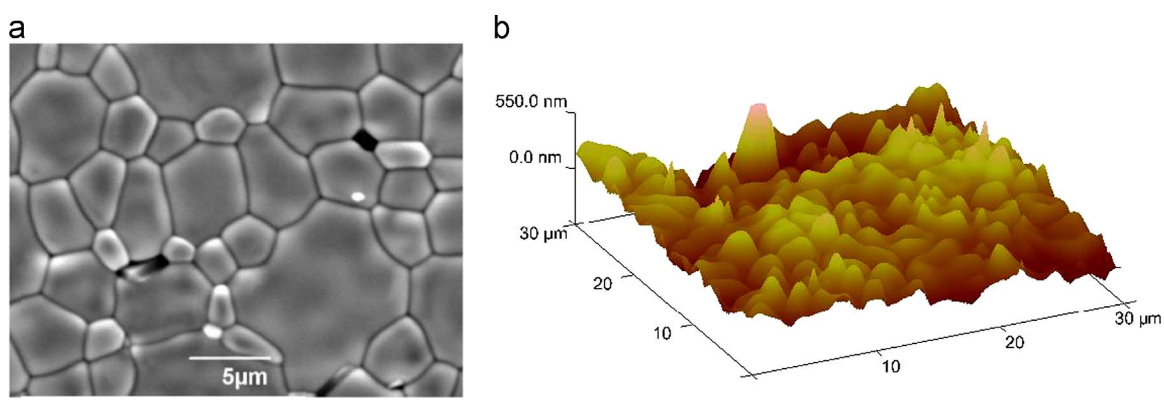

Fig. 1. (a), SEM of LuAG:Ce^{3+} structure, showing grain size about 4 μm; (b) AFM of LuAG:Ce3+ sample on a 30 μm scale, showing roughness of ~100 nm rms.

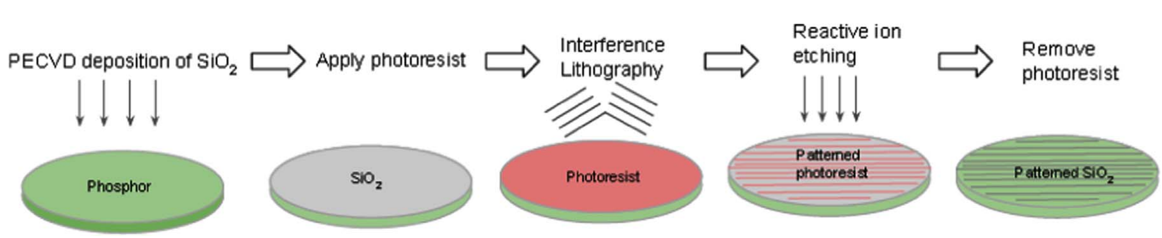

Fig. 2. Overview of SiO_2 grating fabrication process.