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Luminescence spectra of YAG:Ce phosphors synthesized in a field of radiation

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Abstract. The paper presents the measurement results of the luminescence band shape for YAG phosphors upon photoexcitation. It is shown that the position and FWHM does not depend on the excitation technique, but it depends on the phosphor prehistory. The observed difference is assumed to be due to the difference in the structure of nanodefects in phosphors synthesized under different conditions.

1. Introduction

A large number of studies are aimed to develop new synthesis technologies and new compositions to increase the efficiency of phosphors for LEDs. YAG:Ce based phosphors are currently considered the most promising, and many papers are devoted to investigation of these phosphors. The analysis of the published data on spectral characteristics draw attention to distinctions in FWHM and the position of the luminescence bands of phosphors synthesized under different conditions [1-4]. We performed studies of YAG:Ce phosphors of different prehistory to find out the reasons for these distinctions.

2. Experimental

The luminescence spectra were measured for LED lamps and YAG phosphors synthesized at NPO Platan (Russia) and Fultor Enterprises Co. Ltd. (China), and phosphors synthesized in the field of the radiation flux upon excitation by the chip with λ=450 nm and nitrogen laser with λ=337 nm. The study employed SDL3500, SDL4000, YAG 01 and YAG 02 industrial phosphors that obviously differ in technological synthesis modes. The rest of the phosphors were synthesized in the field of the radiation flux [5-7]. Phosphors were obtained by mechanical crushing synthesis and annealed at a temperature of at 1650 °C for 8 hours. After that luminescence spectra of phosphors were measured.

To achieve high measurement accuracy for characteristics of the luminescence bands, a setup was assembled. Figure 1 shows the block diagram of the setup.

A chip with λ=450 nm and a nitrogen laser with λ = 337 nm were used as excitation sources. Both excitation sources excited YAG:Ce phosphors at the maxima of the phosphor absorption bands. The excitation flux was directed to the phosphor. A series of the studied phosphors was placed on a glass substrate in metal washers 1 mm thick with an inner diameter of 6.5 mm.

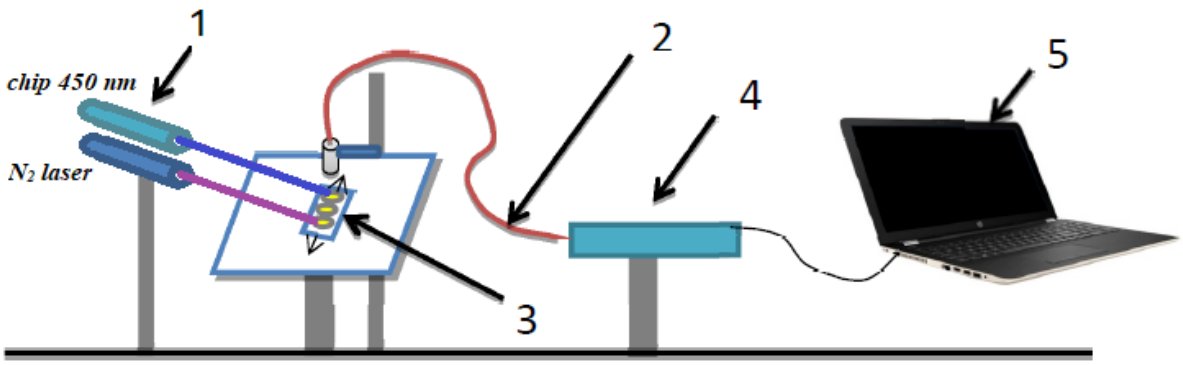

Figure 1. Scheme of the setup to measure the luminescence spectra: excitation sources (1), optical fiber (2), samples on the substrate (3), Avantes AvaSpec-2048L spectrophotometer (4), notebook (5)

The excited luminescence was transmitted through a quartz optical fiber to an Avantes AvaSpec2048L spectrophotometer. The position of the input window of the light guide did not change relative to the sample with the phosphor. A spectrophotometer records the luminescence spectrum, which was then displayed on the laptop display for further processing. The mutual arrangement of all the setup components: the excitation source, the washers with the phosphor, the LED window remained unchanged during the experiment. The measurements were carried out under the same conditions.

During the measurement, the mutual arrangement of the circuit components and the power supply was chosen so as not to distort the shape of emission bands. The position of the band and its FWHM were determined using the WebPlotDigitizer [7] software. The type of data displayed on the monitor is shown in figure 2 as an example.

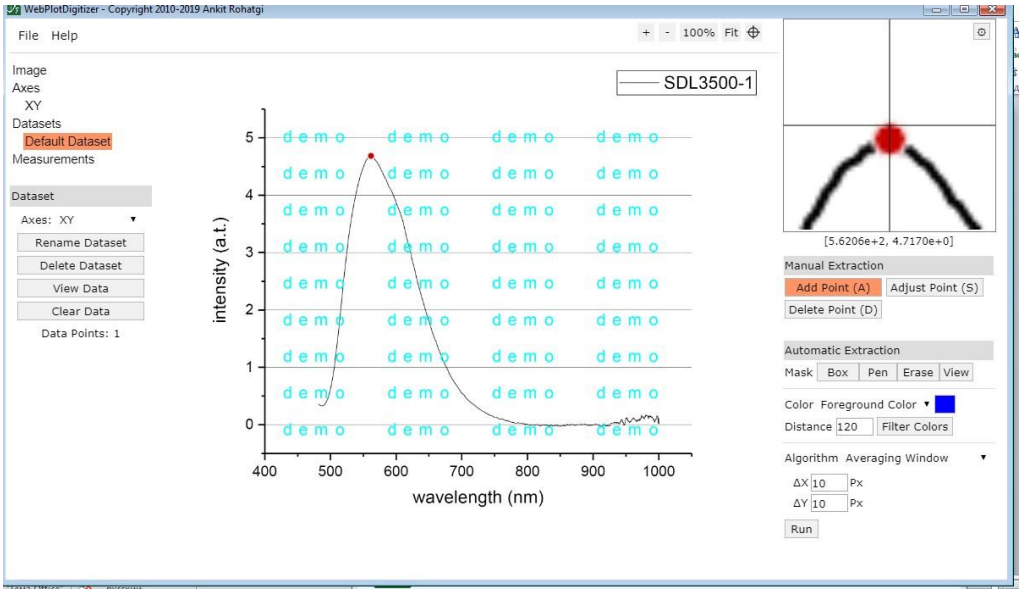

Figure 2. Data in WebPlotDigitizer displayed on the monitor.

3. Results and discussion

In total, 24 types of phosphors were chosen for research. Among them, 4 types of industrial phosphors: SDL3500, SDL4000, YAG 01, YAG 02. The elemental composition of phosphors is summarized in table 1. It was determined using the SEM Quanta3D 200i with XRD system.

Table 1. Elemental composition of the samples of the studied phosphors, in at%

Phosphor	Y	Al	O	Gd	Ce
SDL 3500	13.8	26.0	57.3	2.5	0.4
SDL 4000	13.8	27.0	58.8		0.4
YAG-01	12.5	29.4	54.0	2.7	1.4
YAG-02	12.9	28.0	58.2		0.9

Twenty phosphors were made from ceramics synthesized in the field flow of electron [8]. Luminescence was measured for 10 samples after synthesis and crushing and for 10 samples after subsequent thermal annealing of the same samples at 1650 °C in vacuum. The compositions of 10 ceramic samples are presented in table 2.

Table 2. Elemental composition of the synthesized phosphors

#	**Samples before annealing**
1	$Y_3Al_5O_{12}$: Ce_2O_3(4.8%)
2	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
3	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
4	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
5	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
6	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
7	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
8	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
9	$Y_3Al_5O_{12}$:Ce_2O_3(4%)
10	$Y_3Al_5O_{12}$: Ce_2O_3(1%) + Gd_2O_3(13%)
#	**Samples after annealing**
1	$Y_3Al_5O_{12}$:Ce_2O_3(4.8%)
2	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
3	$Y_3Al_5O_{12}$: Ce_2O_3(2%)
4	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
5	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
6	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
7	$Y_3Al_5O_{12}$:Ce_2O_3(2%) + Gd_2O_3(6%)
8	$Y_3Al_5O_{12}$:Ce_2O_3(2%)
9	$Y_3Al_5O_{12}$:Ce_2O_3(4%)
10	$Y_3Al_5O_{12}$: Ce_2O_3(4.8%) + Gd_2O_3(11.9%)

Phosphors differed in the presence of Ce_2O_3 and gadolinium ions incorpotrated as a modifier. Phosphors 2–4 and 5–7 differed in the prehistory of the starting alumina. The used Al_2O_3 powders were produced in Labor Pharma LLP, Kazakhstan (ChDA TU 6-09-426-75 qualification), Chemical Reagents Plant, Russia (ChDA TU 6-09-426-75) and Hefei Zhonghang Nanotechnology Development Co., Ltd., China (ZH-Al_2O_3-01).

For each type of phosphor, the spectra were measured 10 times, and the statistical measurement error in each case was calculated. The spectrum measurement results: position, FWHM, measurement error are presented in Table 3.

Table 3. Position and FWHM of the luminescence bands in phosphors

#	Type of phosphor	λ_{ex}=337nm		λ_{ex}=450nm	
		λ_{max}, nm	ΔE, eV	λ_{max}, nm	ΔE, eV
		Samples before annealing			
1	1	557±2	0.452±0.005	559±2	0.451±0.005
2	2	538±2	0.446±0.005	537±2	0.455±0.005
3	3	535±2	0.448±0.005	534±2	0.451±0.005
4	4	549±2	0.46±0.005	555±2	0.462±0.005
5	5	548±2	0.452±0.005	552±2	0.448±0.005
6	6	558±2	0.46±0.005	558±2	0.464±0.005
7	7	549±2	0.456±0.005	554±2	0.459±0.005
8	8	557±2	0.449±0.005	558±2	0.450±0.005
9	9	538±2	0.435±0.005	538±2	0.443±0.005
10	10	568±2	0.459±0.005	567±2	0.455±0.005
		Samples after annealing			
11	1	558±2	0.45±0.005	559±2	0.44±0.005
12	2	541±2	0.438±0.005	540±2	0.442±0.005
13	3	538±2	0.455±0.005	535±2	0.445±0.005
14	4	557±2	0.449±0.005	555±2	0.444±0.005
15	5	554±2	0.457±0.005	552±2	0.452±0.005
16	6	556±2	0.469±0.005	557±2	0.463±0.005
17	7	555±2	0.453±0.005	541±2	0.441±0.005
18	8	554±2	0.445±0.005	553±2	0.439±0.005
19	9	546±2	0.437±0.005	544±2	0.440±0.005
20	10	566±2	0.479±0.005	567±2	0.472±0.005
		Industrial phosphors			

21	YAG-01	562±2	0.438±0.005	562±2	0.438±0.005
22	YAG-02	558±2	0.438±0.005	558±2	0.441±0.005
23	SDL-3500	561±2	0.455±0.005	561±2	0.440±0.005
24	SDL-4000	557±2	0.441±0.005	560±2	0.446±0.005

The results obtained show that in all the measured phosphors the band position falls in the range from 535 to 568 nm, and FWHM is 0.435–0.479 nm. For the phosphors synthesized, the band position varies from 535 to 568 nm and FWHM is 0.435–0.479 nm; for industrial phosphors, the band position ranges from 558 to 562 nm, and FWHM is 0.438–0.455 nm.

4. Conclusion

The results obtained in the study of the band shape of phosphors of different prehistory allow us to draw the following conclusions. The positions of the luminescence bands and their FWHM depend on the prehistory, but not on the excitation technique. The difference in the position and FWHM of the bands can be partially attributed to the known effect of the presence of gadolinium. However, the dependence of the band shape in the samples before and after annealing cannot be explained by this effect. Annealing does not change the composition, but it causes ordering of the crystal structure, the environment of the luminescence centers, and cerium ions.

We believe that the difference in the band shape indicates that the structure of the environment of the luminescence centers in YAG:Ce samples of different prehistory is different. This can be due to different technological modes of their synthesis. It is assumed that the observed spread in the luminescence characteristics is caused by the difference in the structure of nanodefects in phosphors synthesized under different conditions [9]. The elemental composition in all nanodefects in YAG: Ce phosphors is similar: matrix ions, dopant, modifier, and intrinsic lattice defects, but their ratio and mutual distribution in the nanodefect is different.

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