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Cathodoluminescence of $Ce:La_2Be_2O_5$ single crystals

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Abstract

$Ce^{3+}:Y_3Al_5O_{12}$ (Ce:YAG) is an important phosphor in high intensity CRT displays since it does not saturate at high electron beam power. The saturation power level is of the order of 1 W/cm^2 for most cathodoluminescent materials, and this limits the maximum surface brightness of a typical cathode ray tube. However, Ce^{3+}:YAG has been found to be linear to at least 10^4 W/cm^2. This performance has encouraged our examination of another cerium activated phosphor, lanthanum beryllate, $Ce:La_2Be_2O_5$ (Ce:BEL). A crystal of lanthanum beryllate activated with a cerium concentration of 410^{18} atoms/cm^3 was grown by the Czochralski crystal growth technique. Wafers were prepared from the crystal and analyzed in both the as-grown state and after annealing in a flowing atmosphere of 10% by volume hydrogen in argon at 1150 °C for 4 h. Cathodoluminescent measurements revealed a broadband blue emission of 100 nm width centered at 480 nm, a blue-shifted analog of the Ce:YAG spectrum. A linear efficiency of 0.13 lm/W was found to a power loading of 8 W/cm^2. Pulsed excitation of a Ce:BEL crystal by 375 nm radiation produced by frequency doubling of a 750 nm laser demonstrated a decay time of the fluorescence of the order of 50 ns. All these data show that $Ce:La_2Be_2O_5$ is an excellent candidate as a blue phosphor for high intensity CRT applications, particularly for high resolution projection displays using single crystal faceplates.

1. Introduction

Conventional CRT faceplates are formed by the deposition of phosphor powder on the inside of a glass envelope of limited thermal conductivity. The image resolution and power capabilities of these faceplates are limited, and many applications now require CRT performance at the limits of phosphor faceplate technology. The resolution of conventional faceplates is limited by phosphor particle size to about 20 μm. Long-term operation at high intensity is limited by a decomposition threshold of about 1 W/cm^2. Short-term operation at high intensity is limited by saturation effects which, aside from thermal quenching and space charge build-up, arise from activator ground state depletion, excited stated absorption and cross-relaxation processes. Activator ground state depletion as a consequence of long fluorescent decay time is a general feature of phosphors [1]. Phosphor particles will actually melt at about 5 W/cm^2. High-intensity operation also limits phosphor lifetime by a process called coulombic degradation. This failure mode reduces the intensity of P53, a standard phosphor, to 50% of its initial value after an electron dosage of 140 C/cm^2. This leads to a CRT lifetime in a high luminance application of about 1000 h under the best conditions.

Single crystal cathode ray tube faceplates have several significant advantages over powder phosphor faceplates. Since single crystals have no granulation, resolution is limited only by the dimension of the electron beam. Single crystal

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phosphor faceplates do not decompose at high power levels, and, because of the intimate thermal contact between the excited phosphor portion of the crystal and the rest of the crystal faceplate, thermal quenching effects are mitigated. Single crystal phosphors have shown no coulombic degradation.

Since the saturation power level is of the order of one watt per square centimeter for most cathodoluminescent materials, this limits the maximum surface brightness of the face of a typical cathode ray tube [2,3]. The single crystal phosphor, cerium activated yttrium aluminum garnet, $Ce^{3+}:Y_3Al_5O_{12}$ or Ce:YAG, is a cathodoluminescent material which has not been found to saturate to an excitation power level of 10^4 W/cm^2 [3,4]. The color of the emitted light from Ce:YAG is yellow-green, and its cathodoluminescent spectrum is sufficiently broad that both green and red colors are available through filtering, but there is practically no light emission in the blue region of wavelength less than 475 nm. Red-shifting of the cerium spectrum by nearly 50 nm has been accomplished by substitution of gadolinium for yttrium, but similar attempts at blue-shifting have been only partially successful [5]. Single crystal cerium-activated yttrium orthosilicate, $Ce:Y_2SiO_5$ or Ce:YSO, a blue phosphor, has been investigated by AT&T Bell Laboratories [6,7] which found saturation at high power.

The performance of Ce:YAG has encouraged our examination of another cerium activated phosphor, lanthanum beryllate, $Ce:La_2Be_2O_5$ (Ce:BEL). BEL is a c-centered monoclinic crystal with lattice constants $a = 0.7536$, $b = 0.7348$ and $c = 0.7439$ nm and $\beta = 91.55°$ [8]. Each unit cell contains four $La_2Be_2O_5$ formula units, and a distinctive feature of the crystal structure is that the lanthanum cations, and the cerium activator cations which substitute for lanthanum, are irregularly coordinated to ten oxygen atoms. The density of lanthanum cations is 1.9410^{22} cm^{-3}, and the refractive indices at a wavelength of 1 μm are $n_x = 1.9641$, $n_y = 1.9974$, and $n_z = 2.0348$. BEL has a melting point of about 1360°C, and a thermal conductivity about half that of YAG.

2. Material preparation

A crystal of cerium activated lanthanum beryllate was grown by the Czochralski crystal growth technique from a heated mixture of cerium oxide, lanthanum oxide, and beryllium oxide in an iridium crucible at 1360°C [9]. The charge consisted of a 330 g total mixture in the following molar percentages: La_2O_3, 49.75%; Ce_2O_3, 0.25%; BeO, 50.00%. This is an effective cerium doping of 0.5% in the melt. Care was taken to correct the weights of these oxide powders for absorbed water. This mixture was fused in a cylindrical iridium crucible of 370 g weight, 2 in (50 mm) in diameter, 2 in (50 mm) in height, using RF heating, and after homogenizing the mixture at a temperature somewhat above 1400 °C, it was cooled to a temperature at which a growth of crystal was observed on the end of a seed crystal of *b*-axis BEL dipped into the molten mixture. A large single crystal, attached to the seed crystal, was then pulled from the melt with 45 rev/min rotation at a rate of 0.5 mm/h in a programmed furnace power cycle which was designed to give a cylindrical crystal shape. After sufficient crystal length had been achieved, the crystal was removed from the melt and slowly cooled. The resultant, approximately cylindrical, crystal of cerium-doped lanthanum beryllate was 163.5 g in weight, 17/4 in (108 mm) long and 3/4 in (19 mm) in diameter.

Mass spectrometric analysis of this crystal showed that the concentration of cerium substituted for lanthanum was 410^{18} atoms/cc. Wafers 19 mm diameter by 3 mm thickness were prepared from this crystal and some were annealed in a flowing atmosphere of 10% by volume hydrogen in argon at 1150 °C for 4 h. It was found that this reduction anneal changed the appearance of the crystal wafers from an orange color to transparent. The reduction anneal also doubled the cathodoluminescent efficiency of the Ce:BEL wafers.

3. Photometry

Wafers were coated on one side with 80 nm of aluminum and mounted in a demountable faceplate cathode ray tube testing station. The wafers

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