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The technique of liquid phase epitaxy has been used to produce single-crystal garnet phosphor screens for cathode-ray tubes. The films were rare-earth-doped yttrium aluminum garnets (YAG's) grown onto YAG substrates using a $PbO:B_2O_3$ flux. We have studied the light output of these layers as a function of film growth temperature, rare-earth activator concentration, and incident power density. With Ce:YAG layers it was possible to use beam power densities up to 10^8 W/m^2 and this produced a radiance of over 10^5 W/cm^2 sr.

PACS numbers: 68.55. + b, 78.60.Hk

One of the limiting factors in obtaining a bright cathode luminescent device is the heat conductivity of the phosphor screen. As the incident electron beam density increases, thermal quenching occurs when the phosphor is heated to a temperature at which the efficiency decreases. Eventually the phosphors "burn out" and become permanently degraded. The actual temperature of the phosphor layer depends not only on the incident beam energy, but also on the manner in which the phosphor screen was fabricated. Normally the luminescent powder is deposited on a glass substrate. Such a structure suffers from both a low heat conductivity of the powder and a poor thermal contact between phosphor and the substrate. The present work was undertaken to overcome these drawbacks in order to obtain higher-brightness cathode ray tubes (CRT's).[1] We have chosen yttrium aluminum garnet ($Y_3Al_5O_{12}$) as a substrate material because it is a good host material for various luminescent ions and because of its high thermal conductivity.

We used the method of liquid phase epitaxy (LPE)[2-4] to grow $Y_3Al_5O_{12}$ layers onto YAG substrates, because this ensured a perfect heat contact between phosphor and the substrate. The solvent was $PbO:B_2O_3$, in which the constituent garnet oxides are dissolved. A typical melt composition used for the growth of YAG was $PbO:B_2O_3:Y_2O_3:Al_2O_3$ $:Re_2O_3 = 450.0:11.65:3.85:6.52:0.02$ (g). This was premelted into a 90-ml Pt/5% Au crucible using a rf generator and then placed into a standard LPE furnace.[3] The activator oxides used in this study were Tb_4O_7, Eu_2O_3, Pr_2O_3, Tm_2O_3, and CeO_2. The substrates were Czochralski-grown YAG, usually [111] and 2.54 cm in diameter.

The efficiency of the layers was determined in a demountable cathode ray tube. The samples were given an Al coating of about 0.08 μm in order to define the energy of the incident electron beam and to carry off the current. The samples were excited by a continuous current of 10 μA at 10 kV in a stationary spot of 4 mm diameter. The light output was measured in transmission. After correction for the energy absorbed in the Al layer (3 kV) and for the refractive index of the phosphor ($n = 1.83$), the measurements yield the internal energy efficiency η. This is only true for films of good quality, i.e., in the absence of surface scattering. In our case this was ensured, because the lattice parameter mismatch between film and substrate was always less than 0.018 Å and

no cracking or facetting occurred.[5]

The chemical composition of the layers was determined by an electron-probe microanalysis where the x-ray line intensities were compared with those for standard materials.

The energy efficiency η can be correlated to the chemical composition of the layers. It appears that both the Pb ions which are incorporated in the garnet lattice from the flux as well as the Re activator ions in the layers influence the efficiency. It is well known from the LPE of iron garnets that the amount of Pb incorporated from the flux increases when the growth temperature is lowered.[6] This holds to a lesser extent for the noniron garnets,[7] and in the present case this is also observed.

The influence of the activator ion concentration is strongly dependent on the Re ion actually used. In Fig. 1 the energy efficiency η is plotted as a function of the concentration per formula unit for [111] YAG films doped with Ce^{3+}, Eu^{3+}, Tb^{3+}, and Tm^{3+}. It is clear that Tm-doped YAG has its maximum at a lower concentration than Eu or Tb. This is in accordance with other work on the concentration quenching of Tm in different host lattices.[8,9] Europium and terbium show an almost identical behavior, except that the overall efficiency of Tb is higher. The cerium ion is a special case. The efficiency rises steeply with concentration, which shows that the Ce ion is a very efficient energy absorber. It
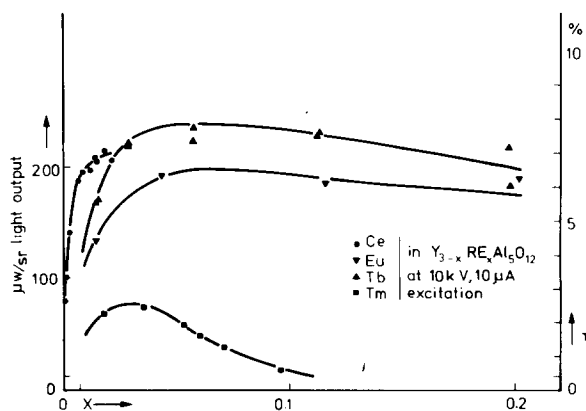

FIG. 1. Light output as a function of the activator ion concentration. The stationary exciting electron beam of 10 μA 10 kV had a diameter of 4 mm.
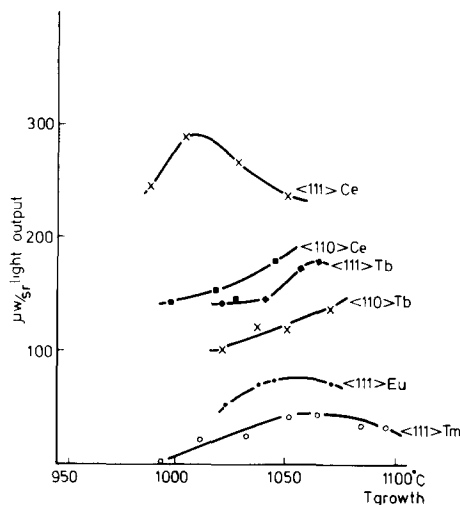
FIG. 2. Light output as a function of growth temperature of [111] and [110] YAG films doped with Ce, Tb, Eu, and Tm. The samples were excited by a stationary electron beam of low energy density.

FIG. 3. Light output as a function of incident power density. The □ refer to measurements in the demountable apparatus and ○ refer to measurements in the electron microprobe.

seems to have a better coupling to the lattice than the other rare-earth ions. The highest measured energy efficiency is higher than that reported by Blasse and Bril[10] for Ce:YAG powder (3.5%). It proved to be impossible to increase the cerium concentration above $x = 0.03$, because at higher concentrations a second phase of CeO_2 appeared in the melt and the incorporation of cerium into the layer did not increase further. A similar effect was observed by Gibbons et al.,[11] who found that the maximum amount of cerium accepted in a garnet powder amounts to 2 mol. % when the powder is prepared under oxidizing conditions.

In Fig. 2 we show the light output as a function of the growth temperature for Ce: Tb: Eu:, and Tm:YAG films. It is clear that the films grown on [111] substrates are more efficient than those grown on [110] substrates for the Ce and Tb activator ions. In the case of [111] films the maxima may be explained by the fact that at low growth temperatures the increase in Pb ion concentration quenches the efficiency, while at the higher growth temperatures the activator ion concentration decreases. These effects are borne out by chemical analysis of the layers.

The lower values of the [110] films shown in Fig. 2 are explained by concentration quenching. Analysis shows that the activator ion concentrations are much higher in these layers than for [111] layers. Thus the maximum in the light output versus growth temperature is shifted to higher values of growth temperature where less activator ion is incorporated. The melt for [110] growth was not further optimized because hillock growth occurs on [110] substrates, analogous to the case of iron garnets explained in great detail by van Erk et al.[12]

In Fig. 3 we show the radiance as a function of the incident power density. At high powers the behavior depends upon the activator ion. The Ce:YAG layers show an almost-linear behavior up to 10^8 W/cm^2, while the Eu:YAG and the Tb:YAG show saturation effects around 10^4
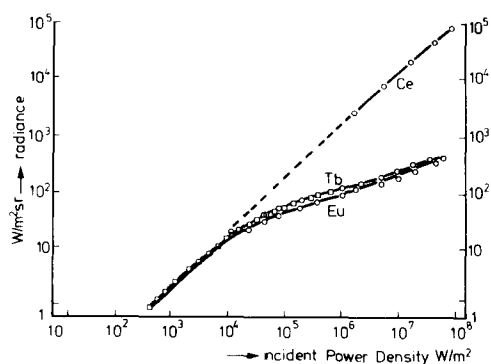
W/cm^2. The temperature of the phosphor at the maximum power density of 10^8 W/m^2 can be calculated[13] to be less than 50 °C. From the known quench temperatures of the phosphors, thermal quenching can be ruled out for Ce, Eu, and Tb. Thulium, with a quench temperature of about 90 °C, remains an intermediate case in this respect. A more detailed explanation of these effects will be given by van der Weg.[14]

In conclusion, we have shown that it is possible to grow cathode luminescent single-crystal thin films by the method of liquid phase epitaxy. These materials have excellent thermal properties so that they can be used with electron beam of high power density without thermal quenching. In particular, the epitaxial layers of Ce:YAG can be used up to power densities of 10^8 W/m^2 and are capable of emitting light with a radiance of over 10^5 W/m^2 Sr.

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