Saturation effects in Y_2SiO_5:Tb under low-voltage excitation

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Low-voltage field emission device phosphors that are excited at high current densities often exhibit brightness saturation with increasing current. The physical processes responsible for saturation can be complex, with several mechanisms contributing, including ground state depletion and excited energy transfer. A two-level model, in conjunction with cathodoluminescence brightness and transient measurements, is used to show the influence of ground state depletion and thermal quenching on the saturation behavior of Y_2SiO_5:Tb under low-voltage excitation. © *2000 American Institute of Physics.* [S0003-6951(00)00908-6]

Recently, Y_2SiO_5 doped with cerium was investigated as an alternative to the standard P22 blue phosphor (ZnS:Ag), currently used in television sets, for low-voltage field emission displays (FEDs).[1] Saturation measurements indicated that the high saturation resistance of the Ce-doped silicate can yield better performance than ZnS:Ag when operated at low voltages (1–3 kV). According to the fast activator theory,[2] fast decay time activators can be used advantageously in FEDs (with a 30 μs pixel dwell time) to reduce ground state depletion and to help overcome saturation. Lee *et al.*[1] have proposed that the superior saturation behavior of the Y_2SiO_5:Ce phosphor can be attributed to the fact that the short decay time of the Ce activator ($\tau_{1/e} \approx 25$ ns) produces a greater resistance to ground state depletion than the slower decay times for ZnS:Ag ($\tau_{1/e} \approx 25$ μs).

To explore the saturation behavior of Y_2SiO_5 in greater detail, transient analysis (measurements of rise and decay times) and brightness measurements were performed under various excitation conditions. The first results for Y_2SiO_5 doped with Tb are presented in this letter.

A Y_2SiO_5 phosphor with 2% terbium concentration was deposited as a deep powder patch and pulsed electron beam excitation was used to measure the luminous efficiency and the cathodoluminescent (CL) rise and decay times for various excitation densities. The CL spectrum obtained at 1 kV and low current densities is shown in Fig. 1. The CL efficiency of this phosphor was measured as 3.8 lumen/W at 1 kV. The transient measurements were made at an acceleration voltage of 1 kV using a 25 ms current pulse and 10 Hz duty cycle to resolve the time dependence. A 72 Hz duty cycle with 30 μs pulse (FED duty cycle) was used for the brightness measurements. The electron beam was pulsed with peak current densities ranging from 1.0 to 300 μA/mm^2.

The saturation behavior of Y_2SiO_5:Tb under low-voltage cathodoluminescence excitation was analyzed using an energy flow model.[3] Using the energy flow model, the external observed energy conversion efficiency η_{CR} can be estimated as[3]

$$\eta_{CR} = (1-\tau_b)\left(\frac{E_P}{\langle \beta_g E_g \rangle}\right)\eta_{tran}\eta_{act}\eta_e, \tag{1}$$

where τ_b is the backscattering coefficient (backscattered primary electrons due to elastic scattering from the ions arranged on the surface layer of the crystal), E_p is the energy of the emitted light, $\langle \beta_g E_g \rangle$ is the average energy required to generate one thermalized electron-hole pair, where E_g is the band gap of the material, and β_g ranges from about 2.7 to 5 depending on the host material,[3] η_{tran} is the transfer efficiency, η_{act} is the activator efficiency, and η_e is the photon escape efficiency (which represents the losses from a phosphor screen). The product of η_{tran} and η_{act} is the light generation efficiency η_{lg} and is a measure of the internal photon generation rate. A simple two level model[2,4] is used to obtain analytical expressions for the transfer efficiency, η_{tran}, and activator efficiency, η_{act}, in Eq. (1). According to this model, the photon emission rate, $d\Phi_{ph}(t)$, during electron beam excitation emitted from a phosphor unit volume element is given by

$$d\Phi_{ph,on}(t) = \frac{\eta_0 g \gamma}{\eta_0 g/N + \gamma_d}[1-\exp(1-\gamma_r t)]dv. \tag{2}$$

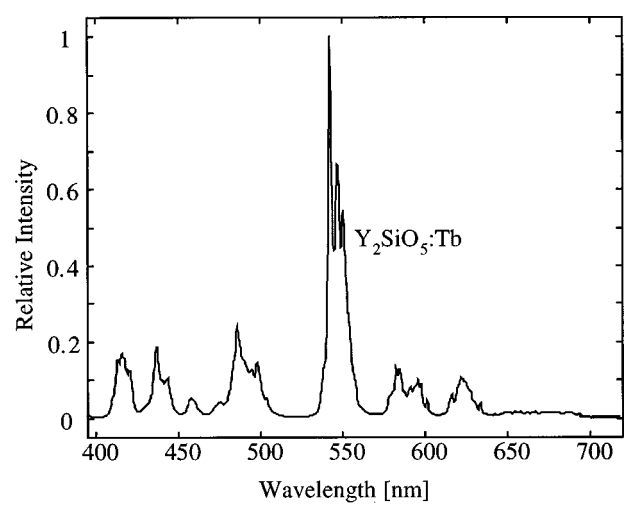

FIG. 1. Luminescence spectrum for Y_2SiO_5:Tb measured at 1 kV.

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Here, η_0 is the transfer efficiency at low excitation densities, g is the generation rate, N is the number of activators, and t is the time after the electron beam has been switched on. The rise rate, γ_r, is given by $\gamma_r = \gamma_d + g(\eta_0/N)$ and the observed decay rate by $\gamma_d = \gamma + \alpha_{\mathrm{act}}$, assuming that g is proportional to the excitation energy density. Here, γ is the radiative decay rate, and α_{act} is the nonradiative decay rate. When the e-beam is turned off after the dwell time Δt, the photon emission rate simply decays according to

$$d\Phi_{\mathrm{ph,off}}(t) = d\Phi_{\mathrm{ph,off}}(\Delta t)\exp(-\gamma_d t). \tag{3}$$

If $\langle \beta_g E_g \rangle$ is the mean energy required for an energetic electron to create an electron-hole pair in a phosphor, the generation factor, i.e., the total number of electron-hole pairs generated per incident beam electron, is given by

$$G = E_b(1-\tau_b)/\langle \beta_g E_g \rangle, \tag{4}$$

where E_b is the electron energy and τ_b represents the fractional electron beam energy loss relating to all of the backscattered and emitted electrons. The local generation rate of carriers, g, can then be written as $g = \langle g \rangle G I_b/e$, where $\langle g \rangle$ is the normalized distribution of the ionization energy in the generation volume, I_b is the electron beam current, and e is the electronic charge. Assuming a cylindrical excitation volume of depth R_e and radius r_b, the normalized distribution function $\langle g \rangle$ can be written as $\langle g \rangle = g(z)/(\tau r_b^2)$.

A universal depth-dose function $g(z)$ was developed from fits to experimental data:[5]

$$g(z) = \lambda_0 + \lambda_1(z/R_e) - \lambda_2(z/R_e)^2 + \lambda_3(z/R_e)^3, \tag{5}$$

with

$$\lambda_0 = 0.60, \quad \lambda_1 = 6.21, \quad \lambda_2 = 12.40, \quad \lambda_3 = 5.69, \tag{6}$$

as calculated by Everhardt and Hoff.[5] Here, z is the depth and $g(z)$ represents e-h pairs generated per unit depth per unit time. This expression represents the number of electron-hole pairs generated per primary electron of energy, E, per unit depth and per unit time.

Using this expression and the range R_e, the energy dissipated at any depth can be determined and hence the carrier-pair generation and light emission in the phosphor can be predicted. The electron-beam energy-dissipation curve for Y_2SiO_5 has been calculated for 1 kV and is shown in Fig. 2. The overall photon emission rate is obtained by integrating Eq. (2) and Eq. (3) over the entire excitation volume. Since each photon of wavelength λ contributes a energy of hc/λ, the total amount of emitted energy is

$$I_{\mathrm{ph}} = \frac{hc}{\lambda}\left[\int_0^{\Delta t}\Phi_{\mathrm{ph,on}}(t)dt + \int_0^{\infty}\Phi_{\mathrm{ph,off}}(t)dt\right]. \tag{7}$$

An important parameter for the effect and magnitude of ground state depletion is the size of the interaction volume. The width of this volume can be approximated by the electron beam spot size, r_b, and the penetration depth R_e, which in turn is inversely related to the acceleration voltage. According to the ground state depletion model, the transfer efficiency should decrease with increasing excitation density. As a result, saturation due to ground state depletion should manifest itself at high current densities and low acceleration voltages.

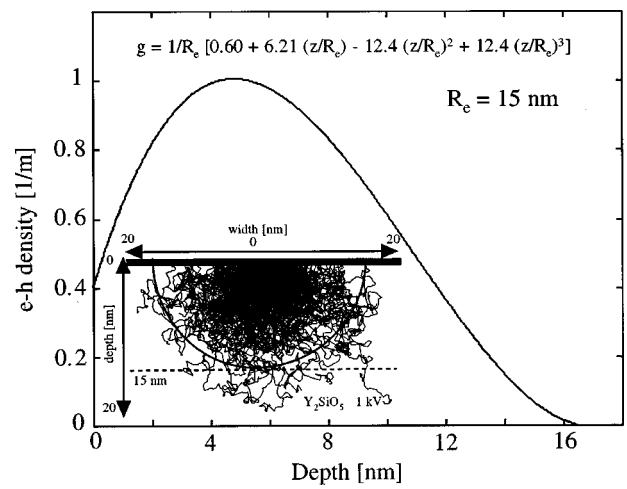

FIG. 2. Generation function for Y_2SiO_5:Tb for a beam energy of 1 keV. Also shown is an electron trajectory plot obtained from Monte Carlo simulations to estimate the penetration depth R.

Since part of the excitation energy is also dissipated in the phosphor material, the electron beam will heat the phosphor. Heating induces a larger nonradiative rate α_{act} and thus γ_d increases with temperature. As a result, the activator efficiency, η_{act}, should decrease with increasing excitation density. Since the transfer efficiency also decreases with increasing excitation density, it is difficult to deconvolute the two effects (thermal effect and ground state depletion). The model presented earlier, however, can be used in conjunction with transient analysis to separate both effects and therefore allows the separation of the impact of ground state depletion and thermal quenching on saturation.

Figure 3 shows three transients for Y_2SiO_5 under different excitation densities, illustrating how the rise time decreases with increasing current density. Clearly, as the peak current density increases from 8.59 to 220 μA/mm^2, the transient times get faster. One possible explanation for this phenomenon is activator ground state depletion.[2] As Eq. (2) suggests, the argument of the exponential term depends on the excitation density and an increase in the excitation density leads to a faster rise of the exponential function. Thus, for a

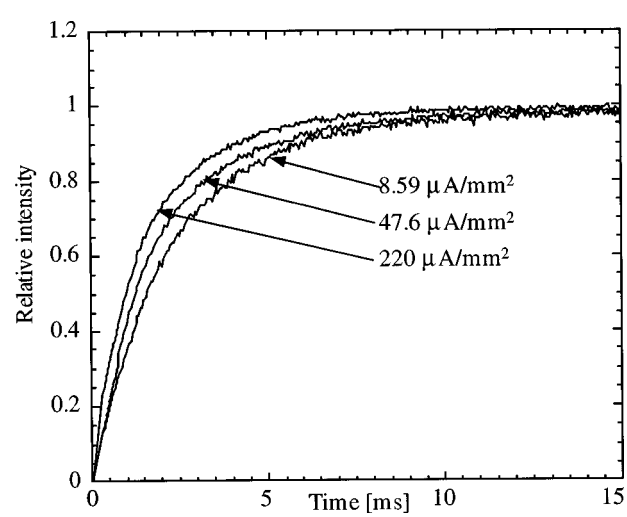

FIG. 3. Cathodoluminescence transients of Y_2SiO_5:Tb with 2% Tb at 1000 volts (25 ms excitation pulse width, 10 Hz repetition rate) for different excitation current densities.

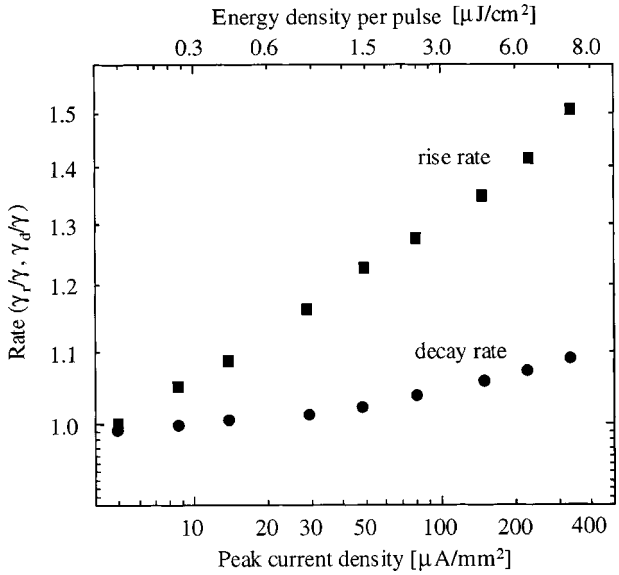

FIG. 4. Rise and decay rates (γ_r, γ_d) at 1000 V (25 ms excitation pulse width, 10 Hz repetition rate) as a function of excitation density per pulse for Y_2SiO_5:Tb.

peak current density of 220 μA/mm^2, the rise of the emission becomes faster than for 47.6 and 8.59 μA/mm^2. This effect is more clearly shown by plotting the rise rate, γ_r, versus the electron beam energy per pulse as shown in Fig. 4. At low excitation densities, when saturation can be neglected, the rise and decay rates are the same. However, with increasing excitation densities, the rise rate will become increasingly faster than the decay rate. The two-level model presented earlier shows that an increasing rate can be attributed to activator ground state depletion. Thus, the increase in the rise rate is an indicator that ground state depletion is present.

Figure 5 shows theoretical curves for the transfer efficiency, η_{tran} as calculated from Eq. (7), Eq. (2), and Eq. (3),

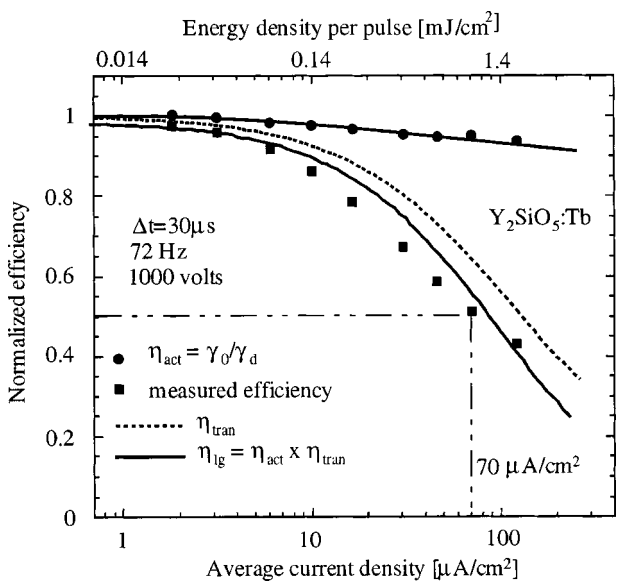

FIG. 5. Efficiency curves for Y_2SiO_5:Tb with 2% Tb at 1000 V as a function of energy density per pulse (30 μs, 72 Hz).

and the light generation efficiency, η_{lg}. Also plotted are the normalized efficiency (squared dots), as measured under FED duty cycle conditions and the activator efficiency, η_{act}, calculated from the change in the decay rate (circular dots). As Fig. 5 suggests, both transfer efficiency and activator efficiency decrease with increasing excitation density. The (transfer) efficiency (without thermal quenching), η_{tran}, is calculated from the ground state depletion model presented earlier. The dissipation profile given in Fig. 2 was calculated from Eq. (5) using a penetration depth of $R_e = 16$ nm, which was calculated from Monte Carlo simulations. As the analysis with the low-level model shows, the transfer efficiency decreases because of the decrease in the available pool of activators. For higher excitation densities, most activators are already in their excited state and the probability of exciting an activator decreases, thus more electron hole pairs recombine through the now more probable nonradiative channels, leading to a decrease in transfer efficiency. The activator efficiency has also a dependence on the excitation density. Here, increasing current density leads to an increase in the energy absorbed and to thermal buildup. Since the probability of the radiative recombination process depends on the temperature, an increase in the excitation density leads to an increase in the energy absorbed, a heating of the phosphor material and, as a consequence, to an increase in the nonradiative rate. This effect can be seen from the slight change of the decay time with increasing current density. However, as Fig. 5 shows, the saturation due to ground state depletion is more pronounced than saturation due to thermal quenching (as the figure suggests, ground state depletion accounts for more than 80% of the observed decrease in efficiency). This is in contrast to saturation phenomena at high acceleration voltages. The higher impact of ground state depletion on the saturation behavior at low voltages can be attributed to the fact that the very shallow penetration (several nm) of electrons at the voltages used in the experiments significantly reduces the volume of available activator. Ground state depletion is more likely to occur at these low voltages than at higher voltages, i.e., 30 kV, where the penetration depth is on the order of several μm.

Transient analysis and brightness measurements can be used to elucidate the different mechanisms for the saturation behavior of low-voltage phosphors like Y_2SiO_5:Tb. Our results show that ground state depletion has a higher impact on the saturation behavior of Y_2SiO_5:Tb than thermal quenching under low-voltage excitation.

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