Electroluminescence thermal quenching in alternating-current thin-film electroluminescent devices

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Abstract

Electroluminescence (EL) thermal quenching in alternating-current thin-film electroluminescent (ACTFEL) devices refers to a reduction in the luminance with increasing temperature, which is in excess to that of normal thermal quenching and is concomitant with a reduction in the transferred charge. A comparison of thermal quenching trends for ZnS:Mn, SrS:Ce, SrS:Cu,Ag, and multi-layer SrS:Cu,Ag /SrS:Ce ACTFEL devices is presented. Respectively, ZnS:Mn; SrS:Cu and SrS:Cu,Ag; and SrS:Ce and multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL devices exhibit very little (normal); a large amount (of primarily EL); or a small amount (of primarily normal) thermal quenching. Insertion of one or more interface layers of SrS:Ce significantly reduces the extent of EL thermal quenching in a SrS:Cu,Ag ACTFEL device. Simulation suggests that SrS:Ce interface layers decrease the rate and displace the location of hole creation by band-to-band impact ionization away from the SrS:Cu,Ag layer, where hole trapping at Cu or Ag acceptor sites leads to EL thermal quenching.
1 INTRODUCTION

The ability to operate over a wide temperature range is an essential advantage of alternating-current thin-film electroluminescent (ACTFEL) flat-panel displays. [1] Thus, the existence of electroluminescence (EL) thermal quenching in blue SrS:Cu and SrS:Cu,Ag ACTFEL devices was considered to be a serious impediment to the commercialization of such phosphors. [2, 3, 4, 5, 6] EL thermal quenching refers to a reduction in the luminance with increasing temperature, which is in excess to that of normal thermal quenching and is concomitant with a reduction in the transferred charge. EL thermal quenching has been attributed to a thermally activated annihilation of positive space charge and a corresponding increase in the threshold voltage. [5]

The purpose of this paper is to present a comparison of EL thermal quenching trends for evaporated ZnS:Mn, atomic layer epitaxy (ALE) SrS:Ce, sputtered SrS:Cu,Ag, and sputtered multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL devices and to demonstrate that the insertion of one or more interface layers of SrS:Ce is an effective means to dramatically reduce the extent of EL thermal quenching. [6] Moreover, simulations are presented which clarify the nature of EL thermal quenching and help to explain why the insertion of SrS:Ce interface layers results in the suppression of EL thermal quenching.

2 EXPERIMENTAL PROCEDURE

The evaporated ZnS:Mn and ALE SrS:Ce ACTFEL devices are similar to those described in reference [7]. The SrS:Cu,Ag single and multi-layer ACTFEL devices employed in this study are fabricated as follows. A SrS:Cu,Ag layer, 1100 nm thick, is deposited by RF sputtering onto a glass substrate coated with layers of indium tin oxide and aluminum-titanium oxide (ATO) which serve as the bottom transparent contact and bottom insulator, respectively. The multi-layer ACTFEL
device containing SrS:Ce interface layers that is reported in this study has a 100 nm SrS:Ce layer between the bottom ATO insulator and a 200 nm SrS:Ce layer deposited on top of the SrS:Cu,Ag layer. The ACTFEL structure is completed with a sputtered barium tantalate oxide insulator (BTO) and Al contacts.

Optical and electrical measurements are performed at 25° C and 50° C for the four types of ACTFEL devices described in the previous paragraph in order to illustrate the range of EL thermal quenching behavior. The substrates are placed in a temperature-controlled environmental chamber (Sun System). Luminance-voltage (L-V), efficiency-voltage (η-V), maximum transferred charge-maximum voltage (Q_{max}-V_{max}), transferred charge derivative-maximum voltage (\frac{dQ_e}{dV_{max}}-V_{max}; this is also denoted transferred charge capacitance), external charge-voltage (Q-V), internal charge-phosphor field (Q-F_p), and capacitance-voltage (C-V) measurements are taken in order to assess the effects of temperature on the threshold voltage (V_{th}), luminance, luminous efficiency, transferred charge, and transferred charge capacitance. [1, 7] L-V and Q_{max}-V_{max} experiments are performed using a 60 Hz bipolar trapezoidal waveform with rise and fall times of 5 μs and a 30 μs pulse width. Q-V, Q-F_p, and C-V measurements are obtained with the bipolar waveform voltage amplitude at 40 V above the 25°C threshold voltage at a frequency of 1 kHz.

3 EXPERIMENTAL RESULTS AND DISCUSSION

3.1 ZnS:Mn and SrS:Ce ACTFEL Devices

We have found evaporated ZnS:Mn ACTFEL devices to perform, in many respects, in a nearly ideal manner. [7, 8] Thus, an examination of the EL thermal quenching tendencies of this type of device can help to establish an ideal performance base-line for EL thermal quenching. Figure 1 shows L-V and η-V curves for an evaporated ZnS:Mn ACTFEL device at 25°C and 50°C. The L-V curves
show that there is only a slight change in $V_{th}$ or the luminous intensity of the ZnS:Mn device over this temperature range. Although the $\eta$-$V$ curves shown in Fig. 1 exhibit a slight decrease with increasing temperature, this decrease is very small. Similarly, the electrical characteristics of this device (not shown) exhibit almost no temperature-dependence. Thus, the electrical and electro-optic properties of evaporated ZnS:Mn ACTFEL devices show very little temperature-dependence between 25°C and 50°C.

Another extensively studied ACTFEL phosphor is SrS:Ce, whose EL thermal quenching properties are also of interest in helping to establish an ideal performance base-line for EL thermal quenching. As shown in Fig. 2, ALE SrS:Ce ACTFEL devices exhibit some temperature-dependence in their L-$V$ and $\eta$-$V$ characteristics between 25°C and 50°C. In the L-$V$ curves shown, the threshold voltage increases slightly with increasing temperature. However, the most dramatic L-$V$ curve trend is the significant decrease in luminance with increasing temperature. This is attributed to normal thermal quenching in which higher temperatures result in an increase in the rate of non-radiative recombination of excited luminescent impurities. Note that this decrease in luminance with increasing temperature is ascribed to normal thermal quenching instead of EL thermal quenching since there is actually a slight increase (not shown) in the transferred charge above threshold at 50°C compared to 25°C. The decrease in luminance and increase in transferred charge combine to reduce the efficiency of SrS:Ce ACTFEL devices as temperature increases. There is also no decrease in overshoot in transferred charge capacitance curves (not shown) in SrS:Ce from 25°C to 50°C. This is evidence that the space charge density is not significantly reduced with increasing temperature in SrS:Ce ACTFEL devices.
3.2 SrS:Cu,Ag ACTFEL Devices

Both SrS:Cu and SrS:Cu,Ag single-layer ACTFEL devices exhibit a significant amount of EL thermal quenching. For example, Fig. 3 shows the L-V and η-V curves at 25°C and 50°C for a sputtered SrS:Cu,Ag ACTFEL device. This figure demonstrates a threshold voltage increase of 14 V and a decrease in luminance of 40%, with increasing temperature; these results are typical for ACTFEL devices fabricated using SrS:Cu or SrS:Cu,Ag phosphors. These temperature trends are attributed to EL thermal quenching since it has been demonstrated that transferred charge and L-V trends correlate quite well. [5] In particular, the threshold voltages estimated from transferred charge and from L-V measurements are found to be very similar and to increase with increasing temperature, resulting in less transferred charge at a given voltage above \( V_{th} \) at higher temperature, as seen in Fig. 4. Note also from Fig. 4 that the above-threshold rise in the transferred charge is less steep for the 50°C curve compared to the 25°C curve and, equivalently, the transferred charge capacitance overshoot is less for the 50°C curve compared to the 25°C curve; both of these trends are consistent with having less positive space charge in the phosphor at elevated temperatures.

Further insight into the temperature-dependence of the SrS:Cu,Ag ACTFEL device is gained from an analysis of \( Q-F_p \) curves, as given in Figs. 5. These \( Q-F_p \) curves are obtained at 40 V above \( V_{th} \) for the threshold voltage at 25°C; in other words, these curves are obtained at the same maximum applied voltage. Note from Fig. 5 that the amount of conduction charge flowing across the phosphor decreases, the polarization charge decreases, and the average phosphor field voltage increases with increasing temperature. An increase in the phosphor field corresponds to a decrease in positive bulk space charge.
3.3 SrS:Cu,Ag ACTFEL Devices with SrS:Ce with Injection Layers

Figure 6 illustrates the L-V and \( \eta \)-V curves at 25°C and 50°C for a sputtered, multi-layer SrS:Cu,Ag ACTFEL device with SrS:Ce interface layers. The significant improvement in EL thermal quenching properties of this device is clearly evident, compared to that shown in Fig. 3 for a single-layer SrS:Cu,Ag ACTFEL device. In particular, the temperature-dependent change in the threshold voltage has been reduced from 14 V to 4 V and the temperature-dependent luminance reduction at +40 V above threshold has been improved from -40% to -14%. Moreover, the introduction of SrS:Ce interface layers results in much less temperature-dependence in the transferred charge characteristics, as shown in Fig. 7; in particular, there is much less change in the threshold voltage and transferred charge capacitance overshoot with temperature. The multi-layer SrS:Cu,Ag device maintains good luminance and luminous efficiency performance with values at +40 V above threshold of 22 cd/m² and 0.2 lm/W, respectively.

Additional confirmation of the significant reduction of EL thermal quenching for multi-layer SrS:Cu,Ag ACTFEL devices with SrS:Ce interface layers is evident from an analysis of the \( Q-F_p \) curves at 25° and 50°C, as shown in Fig. 8; these curves should be compared to those shown in Fig. 5. Although the \( Q-F_p \) curves shown in Fig. 8 change slightly with temperature, these changes are much less pronounced than those observed in Figs. 5.

A comparison of EL thermal quenching trends observed for the four ACTFEL devices considered in this paper (and an additional SrS:Cu ACTFEL device, whose characteristics are not shown in this paper) are summarized in Table I. The first column of Table I is a comparison of the percent reduction in luminance at 40 V above threshold, \( L_{40}\% \). It is evident that evaporated ZnS:Mn ACTFEL devices perform nearly ideally with respect to \( L_{40}\% \), exhibiting the least amount of reduction of all of the devices tested. In contrast, SrS:Cu and SrS:Cu,Ag ACTFEL devices show a very strong reduction in \( L_{40}\% \), while SrS:Ce and multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL
devices are intermediate. Note that a reduction in \(L_{40}\%\) may result from either normal or EL thermal quenching. The last three columns of Table I refer to temperature-dependent electrical properties of the ACTFEL device and thus provide some insight into how much of the thermal quenching is normal and how much is EL thermal quenching in origin; the larger the change in threshold voltage \(V_{th}\), transferred charge at 40 V above threshold \(Q_{40}^{\text{eff}}\), or transferred charge capacitance overshoot (i.e. \(dQ_{\text{max}}/dV_{\text{max}}\) overshoot) with temperature, the larger the contribution of EL thermal quenching. With this in mind, the trend shown in the last three columns of Table I clearly indicated that SrS:Cu and SrS:Cu,Ag ACTFEL devices are dominated by EL thermal quenching, whereas SrS:Ce and multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL devices are dominated by normal thermal quenching. Finally, note from Table I is that single-layer SrS:Cu and SrS:Cu,Ag ACTFEL devices exhibit a large amount of EL thermal quenching, whereas the extent of EL thermal quenching is significantly reduced in the multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device.

3.4 ACTFEL Device Simulations

The n-sheet, state-space ACTFEL device model can be employed to provide insight into the nature of EL thermal quenching. [5, 8] In this model, the ACTFEL device phosphor layer is discretized into \(n\) layers, with space charge modeled as sheet charges between the layers. The hypothesis used to explain EL thermal quenching is that the thermal emission of pre-threshold trapped holes in the phosphor layer reduces the cathodic interface electric field and concomitantly increases the threshold voltage. [5, 8] In this model, positive space charge creation involves band-to-band impact ionization and subsequent hole trapping to a discrete bulk trap; positive space charge annihilation occurs via pure tunneling or thermionic emission from this discrete bulk hole trap. Hole trapping in Cu- or Cu,Ag-doped SrS is expected to be efficient since holes will be coulombically attracted to these negatively ionized \(\text{Cu}_{Sr}\) or \(\text{Ag}_{Sr}\) acceptors.
Parameters of primary interest in the EL thermal quenching simulations are the hole trap density, capture cross-section, energy depth, and the band-to-band impact ionization characteristic field and energy. The hole trap density, in large part, determines the amount of shift in the threshold voltage. This is a simple consequence of the positive trapped charge density correlating with the cathodic electric field enhancement. The capture cross-section also has some bearing on the threshold voltage shift, due primarily to hole trap occupancy considerations, but also in determining the thermionic emission rate of trapped holes. The hole trap depth is set based upon the desired difference in hole emission rates at the temperatures of interest. The hole trap depth is a parameter which establishes the thermionic and tunneling emission rates of trapped holes; choosing a proper trap depth is critical since an excessively shallow trap depth leads to reduced hole occupancies at all temperatures due the domination of tunnel emission (i.e. no EL thermal quenching since hole emission involves tunneling, which is temperature-independent) and an excessively deep trap depth leads to the suppression of thermionic emission (i.e. no EL thermal quenching since thermally-activated hole emission is absent). Finally, the band-to-band impact ionization characteristic field and energy determine the amount of band-to-band impact ionization, and hence, the hole trap occupancy. The values used for these parameters are listed in Table II.

Figure 9 illustrates typical EL thermal quenching results obtained using the n-sheet, state-space ACTFEL device model in the simulation of SrS:Cu or SrS:Cu,Ag ACTFEL devices. The threshold voltage is larger and the above-threshold rise of the transferred charge curve is less steep for the 50°C curve compared to the 25°C curve. Additionally, the transferred charge capacitance overshoot is significantly reduced for the 50°C curve compared to the 25°C curve. These trends are qualitatively similar to the trends shown in Fig. 4.

Figure 10 shows simulated transferred charge and total space charge density curves corresponding to the same situation as for Fig. 9. The total space charge density is the sum of the positive
charge for all of the phosphor layer sheets just prior to the beginning of a positive polarity voltage pulse. Note that the build up of positive space charge begins well below threshold; it then saturates above threshold. The space charge density is always larger for the lower temperature curve, since less thermally-activated space charge annihilation occurs at a lower temperature. It is clearly evident from this figure that positive space charge in the phosphor plays a critical role in establishing the threshold voltage and steepness of the above-threshold rise in transferred charge.

For the 25°C curve of Fig. 10, the space charge density exhibits a distinct step at the threshold voltage. The parameters employed for this simulation are chosen to reveal the existence of this step, which is not always clearly evident in simulation and whose concomitant experimentally-measurable attributes (i.e. an exceedingly steep above-threshold rise in the transferred charge and a large transferred charge capacitance overshoot peak, both of which are seen in the simulated 25°C curve of Fig. 9) are not as dramatic as illustrated in this simulation. This space charge density step is a consequence of the positive feedback-like nature of positive space charge creation by band-to-band impact ionization and subsequent hole trapping; electron tunnel emission from the cathodic phosphor/insulator interface increases greatly with a small increase in the cathode field, resulting in more hole generation, enhancing hole trapping, further increasing the cathodic electric field, resulting in even more electron tunnel emission, thus completing the positive feedback cycle. For the 50°C curve, there is no obvious step in the space charge density at threshold, due to less space charge in the phosphor and concomitantly less positive feedback, so that the above-threshold rise in the transferred charge curve is less steep.

Figure 11 shows simulated transferred charge and total space charge density curves for a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device. This simulation is performed using the same parameters employed in Fig. 10 for the SrS:Cu,Ag portion of the device, with the addition of 200 nm of material at each phosphor/insulator interface in order to simulate the effects of the SrS:Ce layers. The
critical SrS:Ce region simulation issue is to recognize that hole trapping / re-emission, and hence thermally-activated positive space charge annihilation, is not likely to occur in these SrS:Ce regions since positive space charge creation in SrS:Ce appears to involve trap-to-band impact ionization of Ce and native defects. [8] Rather, positive space charge annihilation in these SrS:Ce regions is likely to occur by trapping of electrons injected from the opposite phosphor/insulator interface. Thus, as long as the injection of interface trapped electrons is not thermally-activated, EL thermal quenching is not expected to occur in these SrS:Ce regions. With this in mind, simulation of these SrS:Ce regions is assumed to involve positive space charge creation / annihilation via electron emission / trapping into a discrete donor level 0.9 eV below the conduction band minimum, with a density of $5 \times 10^{16}$ cm$^{-3}$, and a capture cross-section of $1 \times 10^{-14}$ cm$^2$; this is a simple way to simulate non-thermal annihilation of positive space charge.

The most important simulation result presented in Fig. 11 is that EL thermal quenching is greatly reduced by insertion of SrS:Ce interface layers. The threshold voltage increases to about 170 V for both transferred charge curves, as a result of the insertion of the SrS:Ce layers. Below threshold, the total space charge density curves are virtually identical, and then slightly separate above threshold. These trends are a consequence of the fact that hole traps in the simulated SrS:Cu,Ag layer do not fill appreciably until above threshold. Important findings of this EL thermal quenching simulation are: (i) most of the pre-threshold space charge exists in the SrS:Ce layers, not in the SrS:Cu,Ag layer, and (ii) the small amount of above-threshold EL thermal quenching evident in the total space charge density curve of Fig. 11 arises from hole trapping and re-emission in the SrS:Cu,Ag layer. Note that the rate of space charge creation is largest near the cathode phosphor/insulator interface, where the phosphor field is highest. Thus, insertion of SrS:Ce interface layers decreases the rate and displaces the location of hole creation by band-to-band impact ionization away from the SrS:Cu,Ag layer, where hole trapping at Cu or Ag acceptor sites leads to
EL thermal quenching.

4 CONCLUSIONS

A comparison of thermal quenching trends for evaporated ZnS:Mn, ALE SrS:Ce, sputtered SrS:Cu,Ag, and sputtered multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL devices is presented. Evaporated ZnS:Mn ACTFEL devices exhibit the least amount of thermal quenching, which is attributed to normal thermal quenching. SrS:Cu and SrS:Cu,Ag ACTFEL devices possess the greatest amount of thermal quenching, which is primarily EL thermal quenching. SrS:Ce and multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL devices exhibit a small amount of thermal quenching, which is mainly normal thermal quenching. The extent of EL thermal quenching is significantly reduced in a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device, compared to that of a single-layer SrS:Cu or SrS:Cu,Ag ACTFEL device. n-sheet, state-space simulations suggests that the insertion of SrS:Ce interface layers decreases the rate and displaces the location of hole creation by band-to-band impact ionization away from the SrS:Cu,Ag layer, where hole trapping at Cu or Ag acceptor sites leads to EL thermal quenching via thermally-activated annihilation of positive space charge due to thermionic emission of holes from Cu or Ag acceptor traps.

ACKNOWLEDGEMENTS

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References


Table I: Summary of electroluminescence thermal quenching properties for several types of ACTFEL device. Values represent differences between measurements at 25°C and 50°C.

<table>
<thead>
<tr>
<th>Phosphor</th>
<th>$L_{40}$ % change</th>
<th>$V_{th}$ change (V)</th>
<th>$Q_{40}^{crit}$ % change</th>
<th>$\frac{dQ_{max}}{dV_{max}}$ overshoot change</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZnS:Mn (evaporated)</td>
<td>-6</td>
<td>2</td>
<td>-&lt;1</td>
<td>slight decrease</td>
</tr>
<tr>
<td>SrS:Ce</td>
<td>-15</td>
<td>2</td>
<td>+2</td>
<td>no change</td>
</tr>
<tr>
<td>SrS:Cu</td>
<td>-45</td>
<td>10</td>
<td>-12</td>
<td>decreases</td>
</tr>
<tr>
<td>SrS:Cu,Ag</td>
<td>-40</td>
<td>14</td>
<td>-25</td>
<td>decreases</td>
</tr>
<tr>
<td>SrS:Ce/SrS:Cu,Ag/SrS:Ce</td>
<td>-15</td>
<td>4</td>
<td>-1</td>
<td>no change</td>
</tr>
</tbody>
</table>
Table II: n-Sheet, state-space ACTFEL device simulation parameters for thermal quenching analysis. (BBII = Band-to-band impact ionization.)

<table>
<thead>
<tr>
<th>Simulation Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hole Trap Density</td>
<td>$5 \times 10^{16}$ cm$^{-3}$</td>
</tr>
<tr>
<td>Hole Trap Depth</td>
<td>0.65 eV</td>
</tr>
<tr>
<td>Capture Cross Section</td>
<td>$1 \times 10^{-12}$ cm$^2$</td>
</tr>
<tr>
<td>BBII Characteristic Field</td>
<td>2.9 MV/cm</td>
</tr>
<tr>
<td>BBII Ionization Energy</td>
<td>5.4 eV</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Figure 1. Luminance-voltage and efficiency-voltage curves for a ZnS:Mn ACTFEL device at 25°C and 50°C.

Figure 2. Luminance-voltage and efficiency-voltage curves for a SrS:Ce ACTFEL device at 25°C and 50°C.

Figure 3. Luminance-voltage and efficiency-voltage curves for a single-layer SrS:Cu,Ag ACTFEL device at 25°C and 50°C.

Figure 4. Transferred charge and transferred charge capacitance curves for a single-layer SrS:Cu,Ag ACTFEL device at 25°C and 50°C.

Figure 5. Internal charge-phosphor field curves for a single-layer SrS:Cu,Ag ACTFEL device at 25°C and 50°C.

Figure 6. Luminance-voltage and efficiency-voltage curves for a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device at 25°C and 50°C.

Figure 7. Transferred charge and transferred charge capacitance curves for a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device at 25°C and 50°C.

Figure 8. Internal charge-phosphor field curves for a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device at 25°C and 50°C.

Figure 9. Simulated transferred charge and transferred charge capacitance curves for a single-layer SrS:Cu,Ag ACTFEL device exhibiting EL thermal quenching.

Figure 10. Simulated transferred charge and total space charge density curves for a single-layer SrS:Cu,Ag ACTFEL device exhibiting EL thermal quenching.

Figure 11. Simulated transferred charge and total space charge density curves for a multi-layer SrS:Cu,Ag / SrS:Ce ACTFEL device at 25°C and at 50°C. This simulated multi-layer device exhibits reduced EL thermal quenching compared to a single-layer SrS:Cu,Ag device, although the
transferred charge above threshold for the 25°C curve is greater than for the 50°C curve, which is an above-threshold manifestation of EL thermal quenching.
Figure 1: Bankol et al.

Figure 2: Bankol et al.
Figure 3: Bankol et al.

Figure 4: Bankol et al.
Figure 5: Bankol et al.

Figure 6: Bankol et al.
Figure 7: Bankol et al.

Figure 8: Bankol et al.
Figure 9: Baukol et al.

Figure 10: Baukol et al.
Figure 11: Baukol et al.