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Low-voltage driven ZnS:Mn MIS and MISIM thin film electroluminescent devices with Eu_2O_3 insulator layer

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Abstract. AC thin film electroluminescent devices of MIS and MISIM have been fabricated with a novel dielectric layer of Eu_2O_3 as an insulator. The threshold voltage for light emission is found to depend strongly on the frequency of excitation source in these devices. These devices are fabricated with an active layer of ZnS:Mn and a novel dielectric layer of Eu_2O_3 as an insulator. The observed frequency dependence of brightness–voltage characteristics has been explained on the basis of the loss characteristic of the insulator layer. Changes in the threshold voltage and brightness with variation in emitting or insulating film thickness have been investigated in metal–insulator–semiconductor (MIS) structures. It has been found that the decrease in brightness occurring with decreasing ZnS layer thickness can be compensated by an increase in brightness obtained by reducing the insulator thickness. The optimal condition for low threshold voltage and higher stability has been shown to occur when the active layer to insulator thickness ratio lies between one and two.

1. Introduction

Thin film electroluminescent (TFEL) devices with a double insulated structure (Russ and Kennedy 1967) are known for their long life and high brightness. They are emerging as practical flat panel display systems because of a number of attractive properties. These include low power dissipation and the possibility of fabricating large-area solid state multicolour flat panel displays with integrated driving circuits. The most popular double-insulated ZnS:Mn TFEL device makes use of Y_2O_3 as an insulator. Even though they have a high brightness level and long life (Inoguchi *et al* 1974, Inoguchi and Mito 1977) they require a fairly high operating voltage of about 200 V. This makes it difficult to use them with a compact driving circuit composed of available ICs. In some practical systems this problem is solved to some extent by adding an external inductor to the device which is capacitive in nature, thereby forming an LC resonant circuit. Devices with a metal–insulator–semiconductor (MIS) structure have been suggested as one way of reducing the driving voltage requirements of TFEL devices (Kozawaguchi *et al* 1982).

This paper gives details of an AC TFEL device having an insulator with frequency dependent characteristics which modifies the device performance in such a way that it can be switched on and off by changing the excitation frequency while keeping its amplitude constant. This makes high-voltage switching unnecessary and hence minimizes capacitive loading. The device was fabricated with an insulating layer of Eu_2O_3. The choice of Eu_2O_3 as an insulator was motivated by the fact that it has a dielectric constant of 22 and breakdown strength of 2×10^6 V cm^{-1} (Nakane *et al* 1979). However, the loss factor and hence the effective impedence of this material vary with the applied frequency (Jayaraj and Vallabhan 1989a). In order to fabricate low-voltage driven high-brightness stable TFEL devices, MIS as well as MISIM structures have been adopted. This paper reports the detailed emission characteristics of MISIM and MIS TFEL devices using a ZnS:Mn active layer and Eu_2O_3 as an insulator. Optimization of the device parameters is also dealt with in some detail.

2. Device fabrication

The AC TFEL devices with MISIM and MIS structure were fabricated as shown in figure 1. The substrates used were 1 mm thick glass plates. All the layers except the transparent electrodes were deposited by thermal evaporation. The transparent electrodes were prepared by spraying an aqueous solution of $SnCl_4.5H_2O$ with

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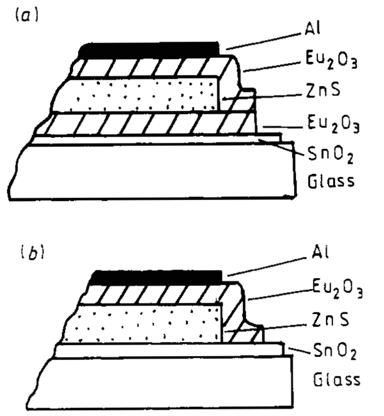

Figure 1. (*a*) Schematic structure of a ZnS:Mn MISIM thin film electroluminescent device. (*b*) Schematic structure of a ZnS:Mn MIS thin film electroluminescent device.

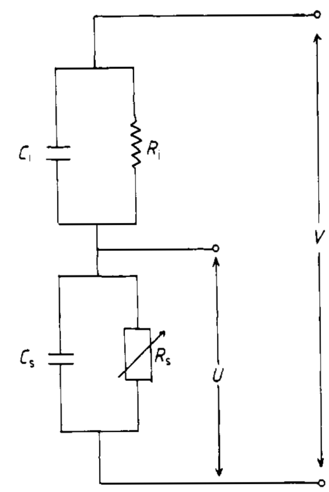

Figure 3. Equivalent circuit.

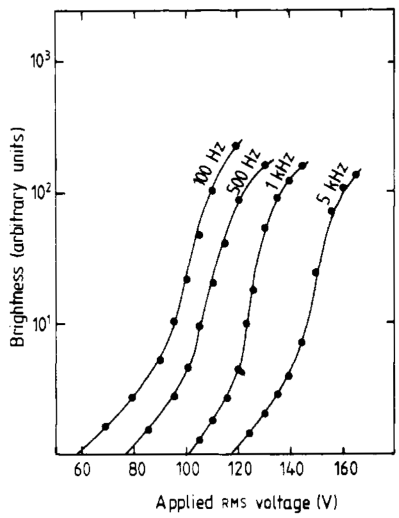

Figure 2. *B–V* characteristics of a ZnS:Mn MISIM structure for different excitation frequencies. Eu_2O_3 = 0.3 μm; ZnS = 0.6 μm.

an excess of isopropyl alcohol to a heated glass substrate kept at 450 °C. Eu_2O_3 films were prepared from pellets of Eu_2O_3 (99.99%) and were deposited by evaporation from tungsten baskets. The active layer was obtained by evaporating electroluminescent ZnS:Mn phosphors prepared in the laboratory by a slurrying technique (Jayaraj and Vallabhan 1989b). During deposition the substrate temperature was maintained at 423 K. The emission characteristics of the device were measured under sinusoidal voltage excitation.

3. Device performance and discussion

The EL emission spectrum and the general nature of the brightness–voltage (*B–V*) curve for the present device are the same as those for the conventional ITO–Y_2O_3–ZnS:Mn–Y_2O_3–Al device (Inoguchi and Mito 1977). From figure 2 it is clear that the driving voltage for the present MISIM structure device is lower when compared with conventional MISIM devices with Y_2O_3 insulators. From the *B–V* characteristics of MIS devices, it is observed that the brightness increases steeply with increasing applied voltage and tends to saturate at higher voltages. The brightness of a typical cell with an active layer of 0.3 μm thickness and insulator 0.2 μm thickness is estimated in absolute units using a PMT having a spectral response closely matching the sensitivity of a human eye (Pillai 1984). Thus, the brightness of the ZnS:Mn device is found to be 1450 ft-lambert ($\approx$4968 cd m^{-2}). However, the *B–V* characteristic obtained for the present device was found to depend strongly on the excitation frequency as can be seen in figure 2. The *B–V* plots clearly show that the threshold voltage for the onset of EL emission increases with excitation frequency. This observation, though anomalous at first sight, can be explained on the basis of the physical properties of the materials used in the fabrication of the device.

The equivalent circuit shown in figure 3 which was used to explain similar behaviour of the *B–V* characteristic of the device with a MgF_2 insulator layer (Pillai and Vallabhan 1986) can also be adopted for the present case. The resistance R_i included in the circuit accounts for the leakage currents. The voltage appearing across the active layer can be expressed as

$$U = \frac{Z_s}{Z_i + Z_s} V \tag{1}$$

where V is the applied voltage, Z_i is the total impedence offered by the insulating layers and Z_s that of the active layer, and C_i and C_s are the equivalent capacitances of the insulator and active layers. If U_{th} is the threshold voltage for the onset of emission and V_{th} is the corresponding applied voltage having a particular frequency ω, then

$$\frac{U_{th}}{V_{th}} = \frac{Z_s(\omega)}{Z_i(\omega) + Z_s(\omega)} = \frac{R_s(1 + \omega C_i R_i)}{(R_s + R_i) + \omega R_s R_i (C_i + C_s)}. \tag{2}$$

From equation (2) it can be seen that U_{th}/V_{th} reduces monotonically with increasing frequency. But devices with other insulators such as Sm_2O_3 and SiO do not exhibit any frequency dependent threshold voltage within the range used here. In the present case the observed effect of variation of threshold voltage for the onset of emission could be due to the presence of Schottky barrier layer formation at the interface of materials of different work functions (Maddocks and Thun 1962). As a result the capacitor C_i can be considered to be a combination of two capacitors in series, one due to the insulating layer and the other due to the interface. Equalization of Fermi levels of the two interface materials (ZnS and Eu_2O_3) results in carrier depletion in the slightly semiconducting dielectric forming the barrier layer. At low frequencies the interface part of the capacitance C_i is independent of dielectric thickness and is very high. At higher frequencies it decreases (Parker and Wasilik 1960) and causes an overall decrease in Z_i. Hence by equation (2) and assuming R_s and R_i to be constants

$$\frac{U_{th}}{V_{th}} \frac{C_i(\omega)}{C_i(\omega) + C_s} \qquad (3)$$

which will exhibit a more pronounced effect in the variation of U_{th}/V_{th}. In equation (3) the capacitance due to ZnS (C_s) is independent of frequency (Maddocks and Thun 1962). An observation which is in support of the above explanation for the occurrence of the frequency dependent threshold voltage is that $\Delta V_{th}/\Delta f$ has been found to be twice as large in a MISIM device as in a MIS device.

The B–V characteristics of MISIM and MIS devices clearly indicate that devices with MIS structures have a much lower threshold voltage compared with MISIM devices. This has prompted us to choose the MIS structure for optimization of the device parameters in terms of active and insulator layer thicknesses.

Figure 4 shows the dependence of brightness on the thickness of the active layer in a ZnS:Mn MIS TFEL device with a constant insulator thickness of 0.2 μm. The brightness was found to decrease as the thickness of the ZnS : Mn active layer was reduced. The threshold voltage of the device also decreased with decreasing active layer thickness. Figure 5 shows the dependence of brightness on the insulator thickness with a constant active layer thickness of 0.3 μm. The threshold voltage again decreases with decrease of insulator layer thickness.

In MIS TFEL devices the mechanism of EL is evidently the impact excitation of emission centres by accelerated charge carriers injected into the active layer. These carriers can originate from the electrodes, ZnS–insulator interfaces, ZnS–SnO_2 interface states and also from the trapping levels in the ZnS layer. The grain size of active layer films has been found to increase with increase of film thickness (Theis 1984). When the ZnS (active layer) thickness is reduced, the number of emission centres correspondingly decreases and the acceleration efficiency is also reduced due to scattering

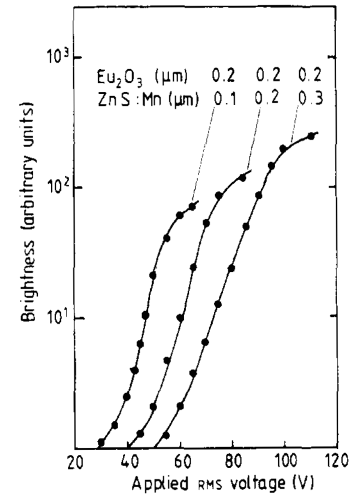

Figure 4. Dependence of brightness on active layer thickness in a MIS structure device with the same insulator thickness, under 1 kHz sinusoidal excitation voltage.

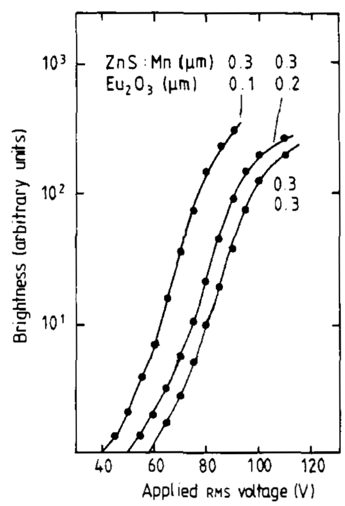

Figure 5. Dependence of brightness on insulator layer thickness in a MIS structure device with the same active layer thickness, under 1 kHz sinusoidal excitation voltage.

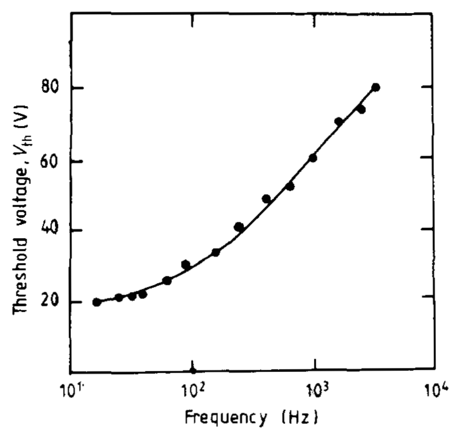

Figure 6. Variation of V_{th} with excitation frequency of a typical TFEL in a MIS structure of active layer thickness 0.3 μm and insulator thickness 0.2 μm.

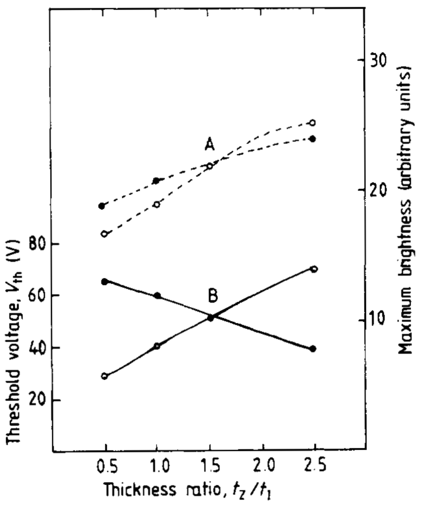

Figure 7. Variation of maximum brightness with t_Z/t_I (curve A): ○ . . . for $t_I = 0.2\ \mu m$; ● . . . for $t_Z = 0.3\ \mu m$. Variation of V_{th} with t_Z/t_I (curve B): ○ —— for $t_I = 0.2\ \mu m$; ● —— for $t_Z = 0.3\ \mu m$.

of electrons at the grain boundaries (Sasakura *et al* 1981, Suyama *et al* 1982). This may reduce the maximum brightness as the thickness of the ZnS layer is decreased. Furthermore, when the active layer thickness is small the number of electrons trapped in the ZnS layer decreases due to electron leakage to the SnO_2 side in the present MIS structure devices. Thus, the number of electrons hot enough to excite Mn centres will be less in a thinner ZnS layer. As the thickness of the active layer is increased the grain size, and the number of Mn centres and trapped electrons, become larger (Theis 1984). These factors influence the maximum intensity which shows a linear rise with an increase in active layer thickness. On reducing the insulator thickness the number of electrons injected from the insulator side is increased. The reduction of insulator thickness thus helps in increasing EL emission intensity. For thicker insulator layers the voltage drop across the insulator is high, necessitating higher applied voltages for making electrons sufficiently hot to excite Mn centres. Hence, a higher applied voltage is required in this case for the onset of emission.

Figure 6 gives the variation of threshold voltage for the onset of emission for a typical cell of MIS structure with an active layer thickness of $0.3\ \mu m$ and insulator layer thickness of $0.2\ \mu m$ for different excitation frequencies.

Figure 7 shows the variation of maximum brightness with the ratio of active layer (ZnS) to insulator thickness (t_Z/t_I). It is found that brightness increases up to a value of 1.5 for t_Z/t_I and then begins to saturate. Curve B shows the variation of threshold voltage with t_Z/t_I at 1 kHz. The threshold voltage is found to increase with increase of either t_Z or t_I. From figure 7 it is clear that a thickness ratio $t_Z/t_I = 1.5$ is best for low-voltage operation.

The driving voltage of TFEL devices can be reduced either by reducing the thickness of the active layer (ZnS) or of the insulator (Eu_2O_3) or both. For a thicker insulator layer $Z_i(\omega)$ is high and hence the voltage drop across the insulator is high causing a decrease in the maximum brightness. When the insulator layer is too thin there is a possibility of breakdown. The optimal condition for high stability as well as brightness is found to be for a thickness ratio lying between one and two for ZnS–Eu_2O_3 layers.

4. Conclusion

A double insulating layer MISIM thin film EL device using a SnO_2–Eu_2O_3–ZnS:Mn–Eu_2O_3–Al structure has been fabricated and compared with a conventional Y_2O_3-based MISIM device. The results clearly indicate that Eu_2O_3 is a better insulator for low-voltage operation. However, the MIS structured device has a much lower threshold voltage than MISIM devices. The *B–V* characteristics of the device show strong dependence on the frequency of the applied voltage. The threshold voltage for emission onset is found to increase with excitation frequency. The *B–V* characteristics of MIS structure TFEL devices are studied in detail. It has been found that the decrease in brightness with decreasing ZnS layer thickness can be compensated for by the increase in brightness obtained by reducing the insulator thickness. The optimal condition for high brightness and stability is obtained for an active layer to insulator layer thickness ratio of between one and two.

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