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To cite this article: Yong Shin Kim *et al* 2003 *Jpn. J. Appl. Phys.* **42** L520

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Low-Voltage dc Thin-Film Electroluminescence with an Indium-Tin-Oxide/CaS:Pb/ZnS/Al Structure

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(Received January 20, 2003; accepted for publication March 26, 2003)

A dc thin-film electroluminescent (TFEL) device has been fabricated to achieve low voltage operation and high-luminance blue emission. The device is based on an [Al/ZnS/luminescent CaS:Pb/indium-tin-oxide] structure in which electrons can be injected into the luminescent layer at high energy due to the field-induced acceleration of electron carriers within the ZnS layer. The blue-emitting CaS:Pb and ZnS layers were consecutively prepared by atomic layer deposition. Preliminary results obtained from this device indicate a threshold voltage for light emission of as low as 17 V and a higher luminous efficiency of 0.1–0.36 lm/W compared with that of CaS:Pb ac TFEL. [DOI: 10.1143/JJAP.42.L520]

KEYWORDS: thin-film electroluminescent device, atomic layer deposition, CaS:Pb, dc electroluminescence, flat panel display

Thin-film electroluminescent (TFEL) devices based on II–VI compound semiconductors have been extensively investigated for their applications to flat panel displays and optoelectronic integrated circuits. To succeed in these applications, it is desired to develop TFEL devices with the characteristics of high luminance and luminous efficiency at low voltage. In particular, lowering the operation voltage is very important to achieve a reduction of power consumption and a simplification of the driving circuitry, which can lead to low-cost fabrication of TFEL panels. The conventional ac TFEL devices with a dual-insulating-layer structure require an applied voltage of between 150 and 250 V to create an electric field (about 1.5 MV/cm) sufficient to discharge electrons from the interface states between the insulating and the phosphor layers and to excite luminescent centers by impact collision.

Many studies have been conducted to achieve low voltage operation using dc TFEL devices.^{1–3)} In these devices, hot electrons are created at low voltage by tunneling through an insulating barrier layer from the cathode to the luminescent layer. Among these, the lattice-matched dc TFEL device fabricated to have a [*n*-Si/CaF₂/ZnS:Mn/CaF₂/indium-tin-oxide] structure¹⁾ is very promising. The device is characterized by supplying hot electrons with high kinetic energy and narrow energy distribution, resulting from the favorable conduction band offsets between *n*-Si, CaF₂ and ZnS:Mn, into the luminescent layer. It lowered the operation voltage to 15 V, which is one order of magnitude less than that of conventional ac TFEL devices. This would increase the luminous efficiency because less energy is required for the hot electrons to excite luminescent centers by impact collision. However, this tunnel device would be difficult to commercialize because of the difficulty of growing of a very thin (2–5 nm) insulating barrier layer over the large area needed for display applications.

In this letter, we report on the blue-emitting CaS:Pb dc TFEL devices prepared by atomic layer deposition. Our device utilized ZnS thin film as the barrier layer between cathodic Al and luminescent CaS:Pb, instead of insulating materials. The ZnS layer prevents the luminescent layer from undergoing an electrical breakdown and gives a chance to supply hot electrons into the luminescent layer by the

field-induced acceleration of electron carriers under a bias voltage. In order to reduce the operation voltage, the thickness of the ZnS layer was chosen to be the minimum thickness, 50 nm, required to accelerate electrons to high energy.⁴⁾

The luminescent region of the dc TFEL device used in the present investigation is a CaS:Pb layer with 250 nm thickness, deposited on an indium-tin-oxide (ITO)-coated glass substrate. The blue-emitting CaS:Pb with a 0.7 at.% Pb concentration was deposited by the controlled atomic layer deposition technique using the precursors of Ca(thd)₂, Pb(C₂H₅)₄, and H₂S at a growth temperature of 390°C.⁵⁾ A pure 50-nm-thick ZnS barrier layer was also grown by surface reactions between diethylzinc and H₂S in the same reactor, consecutively. The thickness and composition of CaS:Pb and ZnS films were confirmed by Rutherford backscattering spectrometry, and their crystal structures were found to be polycrystalline with cubic phases by X-ray diffraction. Al dots with 2 mm diameter were further deposited on the ZnS layer by resistive thermal evaporation in order to provide electrical contacts for luminous and electrical measurements.

Electroluminescent characteristics were measured by applying a unipolar rectangular waveform (100 Hz, 2.5 ms pulse width) to TFEL devices with an ITO electrode having positive polarity. Figure 1 shows the dependence of lumi-

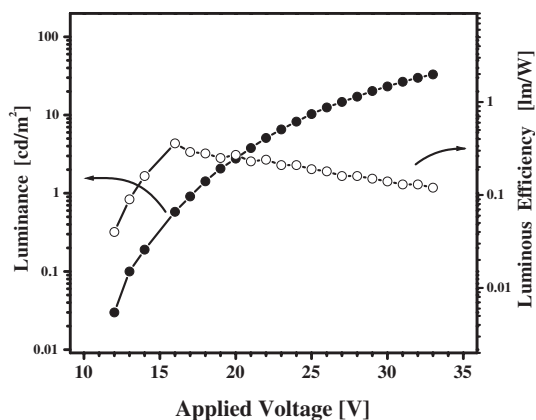


Fig. 1. Luminance and luminous efficiency versus applied voltage for the CaS:Pb device. A unipolar rectangular voltage pulse (100 Hz, 2.5 ms pulse width) was applied to the device with a positive potential at the ITO electrode with respect to Al.

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nance and luminous efficiency on applied voltage for our CaS:Pb dc TFEL device. It exhibited a low threshold voltage of 17 V, and the maximum luminance level was found to be 85 cd/m^2 at 46 V under the same operation conditions. The luminous efficiency had the highest numerical value just below the threshold voltage and then decreased slowly with increasing applied voltage in the range of $0.10\text{--}0.36 \text{ lm/W}$. The luminous efficiency of the CaS:Pb ac TFEL device having the dual-insulating-layer structure was smaller than the value of 0.1 lm/W obtained from separate experiments. Therefore, the higher efficiency suggests that the dc TFEL device is a more advantageous structure for causing electroluminescence compared with the conventional ac device. The light output showed blue emission with a peak at $450\text{--}460 \text{ nm}$ and had a corresponding CIE (Commission Internationale de l'Eclairage) chromaticity coordinate of (0.15, 0.16). Previously, the blue emission had been assigned to the parity-allowed transitions of $6s6p \rightarrow 6s^2$ from Pb^{2+} ions in the dimer center.⁶⁾

The temporal waveforms of light emission intensity $L(t)$ and current density $J(t)$ of the CaS:Pb TFEL device were measured to clarify its operational characteristics. Figure 2 shows $L(t)$ and $J(t)$ together with the applied voltage $V(t)$ using a bipolar triangular waveform at 100 Hz. Discernible current flow and light emission are only observed above an onset voltage when a positive potential is applied to ITO with respect to Al (defined as forward bias). Under a reverse

bias, there is no appreciable current flow, probably due to the suppression of electron injection by the Schottky barrier between ITO and CaS and/or the blocking of carrier transport by the heterojunction barrier between CaS and ZnS. Since the work function of ITO is 0.5 eV higher than that of Al and the band-gap energy of CaS is 0.8 eV higher than that of ZnS, the barrier height for the reverse bias is expected to be larger compared with the forward barrier height. Furthermore, the temporal $J(t)$ and $L(t)$ characteristics were investigated using the trapezoidal applied voltage waveform with rise and fall times of $30 \mu\text{s}$ and a pulse width of $300 \mu\text{s}$. In this case, the rectification behavior for current flow and the turn-on characteristics at the onset voltage were also determined. In addition, current flow and light emission were observed throughout the duration of the applied voltage waveform above the onset voltage. This clearly indicates that the CaS:Pb device operates in the dc mode and that the electrons are continuously injected from the cathodic Al into the CaS:Pb layer.

The current-voltage and luminance-voltage characteristics of the CaS:Pb device for the forward bias are shown in Fig. 3. Both luminance and current show an exponential dependence on applied voltage. The onset voltage for current flow is found to be approximately 16 V, which is identical to the turn-on voltage for light emission. Since the ZnS and CaS layers show a nearly insulating property below the onset voltage due to their high bulk resistivity ($\geq 10^{12} \Omega\cdot\text{cm}$),⁷⁾ the electric field strength corresponding to the onset voltage can be obtained from an equivalent circuit modeled simply with a serial combination of two perfect ZnS and CaS capacitors. The threshold field was calculated to be 0.6 MV/cm . This

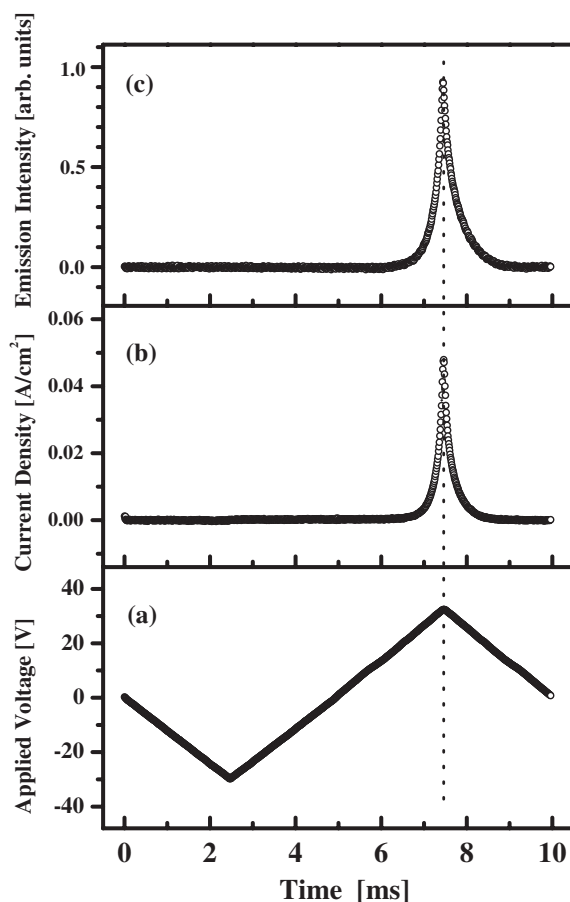


Fig. 2. Time dependence of (a) applied voltage, (b) current density, and (c) light emission in the CaS:Pb device. Forward bias having a positive voltage value is defined as positive potential applied to ITO with respect to Al.

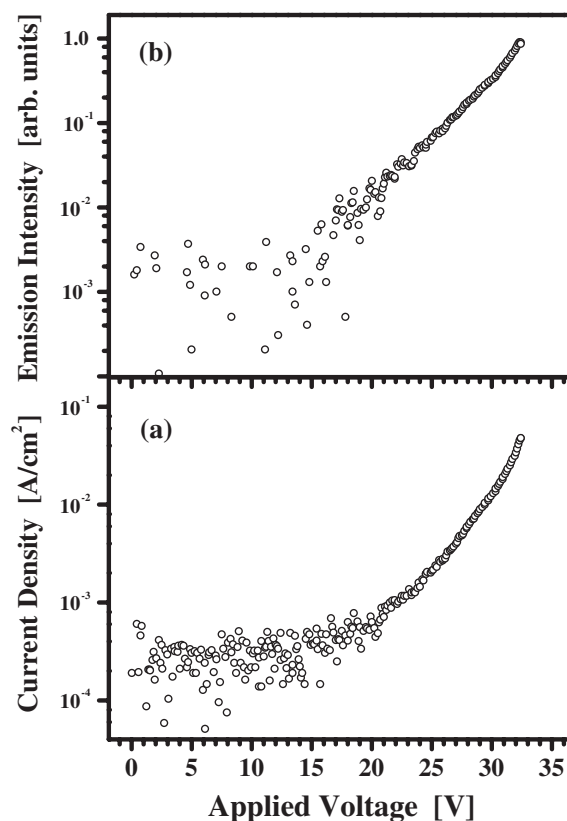


Fig. 3. (a) Current density and (b) emission intensity versus applied voltage for the CaS:Pb device under forward bias.

value is much lower than the electric field strength of 1.5 MV/cm required for conventional ac TFEL devices. Therefore, it implies that hot electrons injected into the active layer have a source different from that of the conventional device, i.e., tunneling from the interface states between the phosphor and the insulating layers. In addition, the Fowler-Nordheim emission from the metal-insulator cathodic contact observed in the previous dc devices seems to be negligible in our dc device because of the relatively thick barrier layer. The ZnS thickness of 50 nm is too large for the wave functions of Al metal to extend into the luminescent layer.

A single CaS device having an [Al/CaS/ITO] structure was found to undergo electrical breakdown easily below the onset field. The current conduction behavior of the dc CaS:Pb device is therefore thought to be mainly determined by the insertion of the ZnS layer between Al and CaS. The current-voltage characteristics of [Al/ZnS/ITO] devices were measured and found to show stable conduction of a significant magnitude in the high-field range of 0.5–2.0 MV/cm under forward bias, as observed in the CaS:Pb dc TFEL device. Such high-field conduction in the single ZnS device was previously reported and explained in terms of the bulk-limited rather than the interface-limited mechanism,⁸⁾ even though the Al/ZnS barrier height had been observed to be 0.8 eV in a very clean system.⁹⁾ The discrepancy had been explained by the extreme sensitivity of the interface properties to defects. Consequently, the bulk-limited conduction in the ZnS layer is inferred to be the current-controlling factor underlying the operation of the CaS:Pb dc TFEL device.

Four striking features of the CaS:Pb dc TFEL device, as mentioned above, are summarized as follows: (i) a higher luminous efficiency compared with the corresponding ac TFEL device, (ii) a continuous injection of hot electrons into the luminescent layer, (iii) a lower onset field strength (0.6 MV/cm) for light emission, and (iv) a current regulation by the ZnS layer. Considering these features, we propose the following electroluminescence mechanism for our device. Electrons are easily injected from the Al cathode electrode into the ZnS layer probably through interface defect states

under forward bias. The amount of injected electrons is mainly controlled by the bulk-limited mechanism in the ZnS layer having a significant conduction above an onset voltage. The injected electrons are accelerated by the applied electric field and obtain kinetic energy within the ZnS layer. The maximum energy gained at the onset field is calculated to be 3 eV. The hot electrons with high kinetic energy enter into the CaS:Pb luminescent layer and are then further accelerated within the CaS layer. During the passage through CaS:Pb layer, impact collisions between the hot electrons and the Pb^{+2} dimer centers result in the blue emission. The high luminous efficiency and the low onset field strength are well explained by the injection of hot electrons with high kinetic energy into the luminescent layer.

In conclusion, we have demonstrated a new low-voltage dc TFEL device fabricated by the successive growth of blue-emitting CaS:Pb and barrier ZnS layers by atomic layer deposition. The use of ZnS as an electron-accelerating and current-controlling layer has made it possible to operate the device at a low voltage with reliable stability, by providing a suitable amount of hot electrons with high kinetic energy into the luminescent layer. This device has optical turn-on voltage reduced by one order of magnitude and higher luminous efficiency compared with conventional ac TFEL devices.

This work was financially supported by the Korea Ministry of Information and Communications.

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