Roughness of $ZnS:Pr,Ce/Ta_2O_5$ Interface and Its Effects on Electrical Performance of Alternating Current Thin-Film Electroluminescent Devices

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***Abstract*—Roughness effects of neighboring dielectrics on electrical characteristics of thin-film electroluminescent devices were investigated in order to improve the understanding of physics for the devices. Atomic force microscopy analysis reveal that thicker bottom layer of Ta_2O_5 shows rougher surface resulting in the rougher surface of $ZnS:Pr,Ce$ layer. It can be easily seen that the dc leakage current increases rapidly with increase of surface roughness Furthermore, it is notable that the initiation field of Poole–Frenkel current conduction is lowered by increasing surface roughness of Ta_2O_5 thin film. Internal charge-phosphor field ($Q_{int} - F_p$) analysis and capacitance-ac voltage (C–V) analysis for ITO-Ta_2O_5-$ZnS:Pr,Ce$-Al and ITO-Ta_2O_5-$ZnS:Pr,Ce$-Ta_2O_5-Al show that the steady state phosphor field is smaller and C–V curve in transition region is less steep with increase of root-mean-square roughness between lower dielectric and phosphor layer in the alternating current thin-film electroluminescent (ACTFEL) devices. Therefore, we conclude that interface roughness is one of the physical factors to change the electrical performance of ACTFEL device.**

***Index Terms*— Electroluminescence, insulator, interface, Ta_2O_5, TFEL, ZnS.**

I. Introduction

THE alternating current thin-film electroluminescent (ACTFEL) devices are of scientific interest since they offer an useful mean of studying the physics of insulator-semiconductor interfaces. Several reports [1]–[3] have analyzed the relation between insulator-semiconductor interface and the characteristics of ACTFEL devices. The ac bias applied across the device acts to alternately accelerate the electrons from one insulator-semiconductor interface to the other. Light generation in the phosphor layer is attributed to impact excitation of activator centers by high-energy electrons [1]–[3]. It is reported that the variation of surface roughness plays an important role in the performance of insulating film [4]. In the same context, it is expected that roughness would act as an important physical parameter for the performance of ACTFEL devices where the properties of the insulator and phosphor interface have an important effect on the operating characteristics such as turn-on, reliability, and degradation, etc.

Recently, Singh *et al.* proposed that dielectric layer roughness has an effect on the ACTFEL device characteristics, especially current crowding [3]. In this study, we have investigated the effect of the surface roughness on dc or ac electrical characteristics of MIM(ITO/Ta_2O_5/Al) capacitor and ACTFEL devices with MISM(ITO/Ta_2O_5/$ZnS:Pr,Ce$/Al) structure and ACTFEL devices with MISIM(ITO/Ta_2O_5/$ZnS:Pr,Ce$/Ta_2O_5/Al) structure. The measured characteristics are explained in terms of the surface roughness obtained by atomic force microscopy. Our results indicate that the interface roughness between phosphor and lower insulating film strongly influence on interface characteristics as well as the internal parameters such as phosphor field.

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II. Experiments

To this end, M(ITO)-I(insulator)-M(Al) structures using Ta_2O_5 film and ACTFEL devices with M-I-S(Phosphor)-M and MISIM structure were fabricated. The Ta_2O_5 layer as a bottom insulator was deposited using a conventional rf-magnetron sputtering technique at substrate temperature of 200 °C using the Ta_2O_5 target (4 in diameter) with 99.99% purity on the 7059 glass coated with the transparent electrode ITO(Sn-doped In_2O_3) with a sheet resistance of about 20 $\Omega/\square$. The base pressure in the chamber was adjusted to 3×10^{-6} torr and the pressure during the deposition was maintained at 10 mtorr of Ar (80%) and O_2 (20%) gas mixture to suppress the formation of oxygen defects which may become electron traps in the film.

In order to study effect of the thickness on the roughness of Ta_2O_5, the thickness of lower Ta_2O_5 was varied to 200 nm (#1), 300 nm (#2), and 400 nm (#3). The dielectric constant and dielectric loss ($\tan \delta$) for the Ta_2O_5 films formed in the present studies were 22–26 and 0.2–0.6%, respectively, which were determined using HP 4192A impedance analyzer. Subsequently, white light-emitting $ZnS:Pr$ (0.3 mol%), Ce (0.3 mol%) phosphor layer was deposited by electron-beam evaporation method. After the deposition of the $ZnS:Pr,Ce$, the film was annealed at 450° for half an hour in vacuum (3×10^{-6} torr) for the crystallization of host matrix and an efficient diffusion of the activators. Then, aluminum was thermally evaporated on the top of $ZnS:Pr,Ce$ layer to fabricate ACTFEL devices with M-I-S-M structure. The EL devices with MISIM structure were fabricated by the deposition of the upper Ta_2O_5 on the top of the $ZnS:Pr,Ce$ layer. For electrical measurements, aluminum top electrodes of 0.7 mm in diameter and 100 nm in thickness were thermally evaporated through a shadow mask.

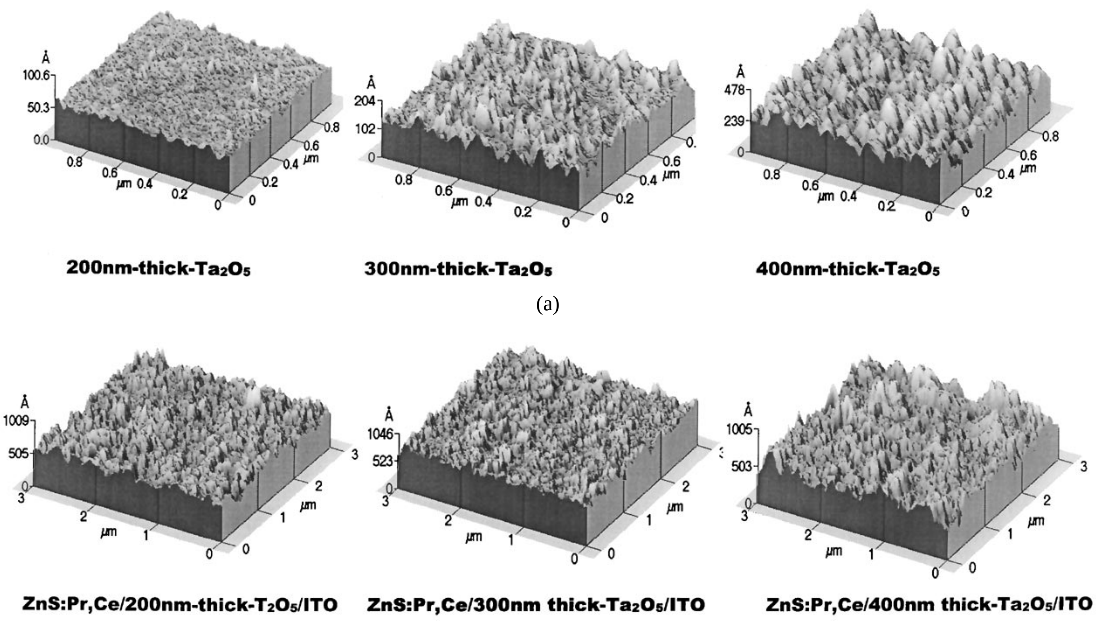


Fig. 1. (a) AFM images for the Ta_2O_5 surface with the variation of deposition thickness; 200 nm, 300 nm, and 400 nm in thick and (b) surface roughness of ZnS : Pr,Ce layer on 200 nm Ta_2O_5, ZnS : Pr,Ce layer on 300 nm Ta_2O_5 layer, ZnS : Pr,Ce layer on 400 nm Ta_2O_5 from the left.

In order to examine conduction mechanism and properties for the Ta_2O_5 and EL devices with MISM structure we measured dc current density–voltage characteristics (J–V) at room temperature with Keithely 237 source/measure unit. The ac analysis (internal phosphor charge-internal phosphor field ($Q_{\mathrm{int}} - F_p$) and capacitance–voltage (C–V) analysis) for the TFEL devices were accomplished using a conventional test circuit [5]–[7]. The standard driving waveform employed was obtained using an arbitrary waveform generator (HP 33 120A) in conjunction with high power linear amplifier and was symmetric with alternating bipolar pulses of trapezoidal shape with 40 μs rise and fall times and a pulse width 80 μs.

III. Results and Discussion

Fig. 1(a) shows the change of surface roughness of Ta_2O_5 films observed by atomic force microscopy (AFM) analysis with increase of thickness. The peak on the uneven growing surface receive more incident flux than valley and nucleus growth at the valley decrease in rf sputtered insulator. So, the roughness increases with increasing thickness of Ta_2O_5 layer. On the other hand, to investigate the effect of surface roughness of lower Ta_2O_5 and the interface roughness between ZnS : Pr,Ce and Ta_2O_5 layer on dc electrical characteristics, the thickness of both ZnS : Pr,Ce layer and upper Ta_2O_5 layer for all ACTFEL devices was fixed at about 300 nm. The root-mean-square roughness of ZnS : Pr,Ce on 200 and 300 nm Ta_2O_5 layers is about 70 Å but that of 400 nm one is 90 Å. We confirmed that the roughness of ZnS : Pr,Ce tend to increase with increase of roughness of bottom insulator, as shown in Fig. 1(b). The increase of surface roughness of ZnS : Pr,Ce might be caused by rougher surface of bottom insulator than intrinsic growth characteristics of ZnS film.

Fig. 2 shows dc leakage current density of Ta_2O_5 capacitors as a function of applied electric field. As shown in Fig. 2, it

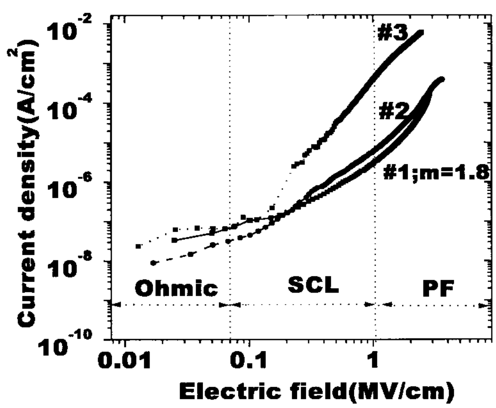


Fig. 2. Leakage current densities of ITO/ Ta_2O_5/Al capacitors as a function of electric field. The currents increase with increasing surface roughness of Ta_2O_5 layer.

can be easily seen that the current density increases rapidly with increase of surface roughness. At low electric field, the Ta_2O_5 films typically exhibit a linear ohmic conduction. The nonlinear behavior of the leakage current at the electric field exceeding about 0.1 MV/cm may be governed by the space charge limited (SCL) conduction process with a relationship of $J \propto E^{1.8\sim2.3}$ at middle field region. The sample #1 and #2 show similar characteristics of current density-electric field. At the electric fields of order of 1 MV/cm or more, the Poole–Frenkel emission are the most obvious mechanisms.

In Fig. 3, the current density-electric field data of Fig. 2 are plotted for three Ta_2O_5 films as $\ln(J/E)$ versus $E^{1/2}$ (Poole–Frenkel plot) at high field region. From the slopes of the linear region of Fig. 3, a high-frequency dielectric constant of 4.4–9 ($\varepsilon = n^2$: n = refractive index) was estimated for the films and the value is in agreement with the earlier reported values [8]–[10]. Furthermore, it is notable that the

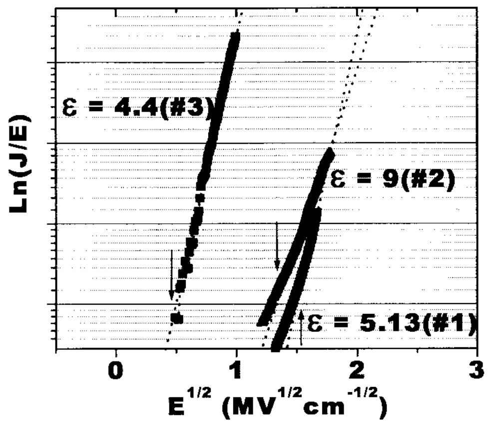
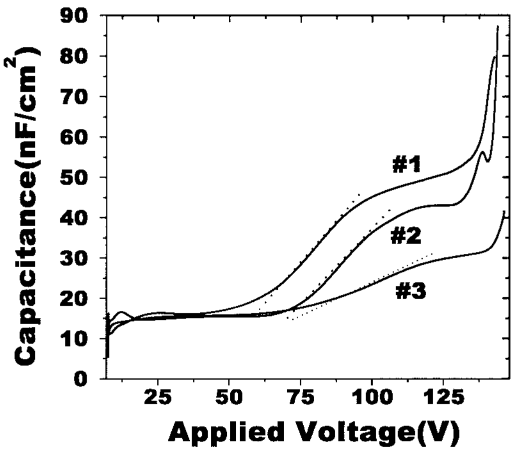


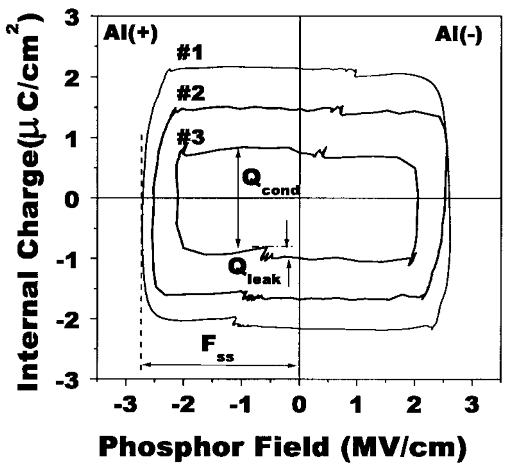
Fig. 3. Poole–Frenkel plots for the leakage current of ITO/Ta_2O_5/Al capacitors in high field region. The arrow indexes indicate the initiation field of Poole–Frenkel conduction process.

initiation field of Poole–Frenkel current conduction is lowered by increasing surface roughness of Ta_2O_5 thin film. Thus, the current of #3 appears to be more strongly governed by the P–F mechanism even at low field than in #1 and #2. Since the P–F emission is due to the field-enhanced thermal excitation of trapped carriers, we assume that #3 are richer in defects than #1 and #2. These results indicate that the leakage current level under dc high field show different behaviors depending on roughness of TO and the enhanced roughness may results in a large amount of defects at and near surface as well as lowering onset field for conduction, even if the deposition conditions are same. Although the leakage current level is higher than that of conventional Si-based insulating films, leakage currents less than about 1 mA/cm^2(=1 μC/cm^2/1 kHz) at all Ta_2O_5 capacitor under the electric field of 2 MV/cm could be achieved. This value permits sufficient margins for ACTFEL devices operation.

The observed C–V and $Q_{\rm int} - F_p$ loops at $V_{\rm th+40}$ are shown in Fig. 4(a) and (b) for the TFEL devices with MISIM structure. A family of C–V curves is presented as a function of the surface roughness of lower Ta_2O_5 layer in Fig. 4(a). At Al(+), note those C–V curves in transition region are less steep with increase of surface roughness. This result indicates that if the thickness of Ta_2O_5 is fixed at same value, the density of interface state in the preclamping field regime is larger as interface roughness of ZnS : Pr,Ce and Ta_2O_5 layer is rougher. From the $Q_{\rm int}$–F_p characteristics as shown in Fig. 4(b), it should be noted that the steady state phosphor field ($F_{\rm ss}$) and turn-on field is lower with increasing surface roughness of lower Ta_2O_5 layer at Al(+). Furthermore, the ACTFEL device with higher roughness of ZnS : Pr,Ce has lower steady state phosphor field when Al electrode is negatively biased. Assuming that the sharp point due to rougher surface easily generate tunneling electron at the low field and the internal polarization is built up by the stored charge at the opposite interface, the resulting internal voltage will then be decreased as compared to that of #1 and #2. Also, the maximum voltage applied to the ZnS layer which is given by $V_{\max} = E \bullet d_{\rm ZnS}$ where E is the clamped field strength should be limited at the low value by the weakened area of sharp points. Thus, it is



(a)



(b)

Fig. 4. (a) C–V curves for ACTFEL devices with MISIM structure which have lower Ta_2O_5 layer of 200 nm (#1), 300 nm (#2), and 400 nm (#3)-thick and (b) $Q_{\rm int}$–F_p for ACTFEL devices with MISIM structure which have lower Ta_2O_5 layers of 200 nm (#1)-, 300 nm (#2)-, and 400 nm (#3)-thick.

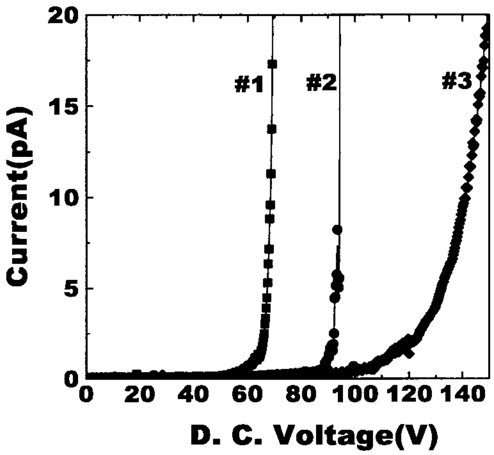
natural that the maximum steady state field would be limited in the fixed range.

Some supporting evidences for these are observed in dc I–V characteristics in ACTFEL devices with MISM structure. As displayed in Fig. 5(a) and (b), turn-on voltages of Fowler–Nordeim (F–N) tunneling for the sample #2 and #3 are nearly same, as shown in enlarged figure, though slope is remarkably different and also, this is in line with Figs. 2. and 4(a). Sample #1 and #2 show typical Flowler–Nordheim conduction under the high field above 1 MV/cm while the conduction mechanism of sample #3 is deviated from the behavior for #1 and #2. This plot indicate that trap depth and distribution of sample #3 are different from those of sample #1 and #2. Though it is very tentative, there is a sufficient possibility that tunneling initiate from ITO electrode to Al via bulk of Ta_2O_5.

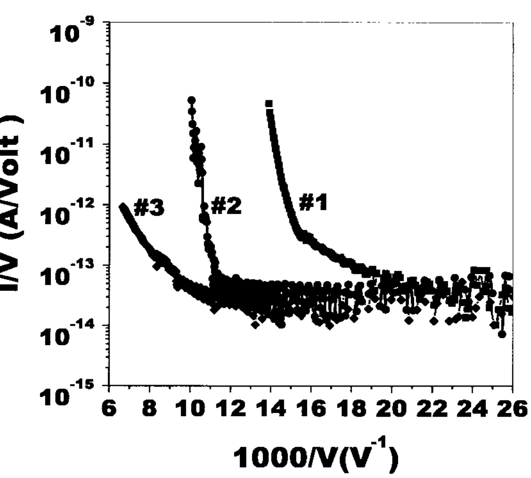
The steady state phosphor field characteristics of ACTFEL devices with MISM structure in Fig. 6(a) show similar result with those of ACTFEL devices with MISIM structure. The relations between steady state phosphor field of ACTFEL devices with MISIM structure and the surface roughness of

TABLE I
A COMPARISON OF UPPER AND LOWER INTERFACE ROUGHNESS AND STEADY STATE ELECTRIC FIELDS OF AC TFEL DEVICES WITH MISIM STRUCTURE

MISIM structures \ Parameters	RMS Roughness of lower Ta_2O_5	RMS Roughness of ZnS:Pr,Ce /Ta_2O_5	F_{ss}^{+} (MV/cm)	F_{ss}^{-} (MV/cm)	Q_{leak} /Q_{cond}
200/300/300nm(#1)	0.4nm	7.6nm	2.7	2.5	0.026
300/300/300nm(#2)	1.7nm	7.1nm	2.5	2.5	0.028
400/300/300nm(#3)	5.0nm	9.0nm	2.1	2.0	0.071

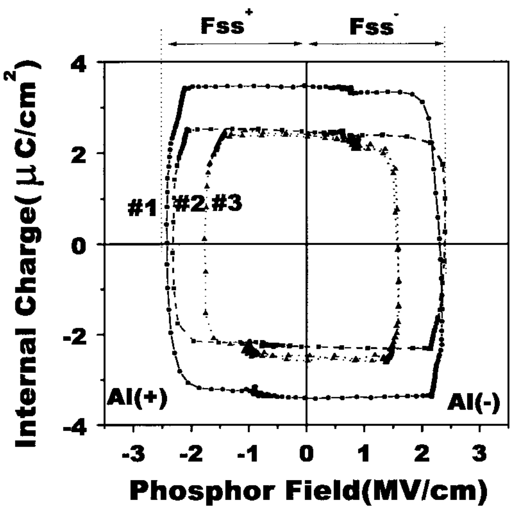


(a)

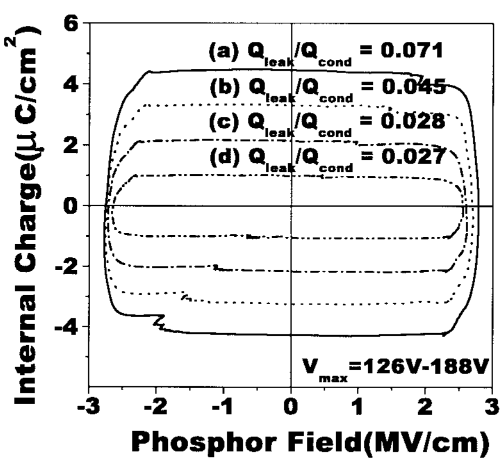


(b)

Fig. 5. DC $I-V$ characteristics and Flowler–Nordheim plot for TFEL devices with MISM structure when Al electrode was positively biased: (a) DC $I-V$ characteristics and (b) Fowler–Nordheim plot for curve (a).



(a)



(b)

Fig. 6. (a) $Q_{\rm int}$–F_p curves for ACTFEL devices with MISM structure which have lower Ta_2O_5 layers of 200 nm (#1), 300 nm (#2), and 400 nm (#3) thickness. (b) $Q_{\rm int}$–F_p curves as a function of maximum applied voltage ($V_{\max}$) for #2 TFEL device. $V_{\max}$ were varied in the rage of 126–188 $V_{\rm peak}$.

Ta_2O_5 layers and ZnS : Pr,Ce layers are summarized in Table I. Superscripts of + and − used in Table I have conventional meaning of the polarity of the applied voltage pulse. The observed results are summarized as follows: 1) the rougher dielectric-phosphor interface can be expected to have relatively many sharp peaks where the electric field would be higher than smoother one, and 2) with increasing roughness of dielectric-phosphor interface, the lower turn-on field and the lower steady state phosphor field is resulted.

A series of $Q_{\rm int}$–F_p curves is shown as a function of applied voltage ($V_{\max}$) in Fig. 6(b). It is noted that the steady state phosphor field at Al(+) and Al(−) are not so much depended on $V_{\max}$ while the fraction of leakage to conduction charge increases with increase of $V_{\max}$. Although the phosphor field of sample #3 is smaller than the other samples, the fraction of leakage to conduction charge is much larger than the others. It reveals that much of the charge transported across the phosphor of sample #3 appears to reside in relatively shallow traps such that it can easily emit from these traps when the external bias is zeroed. If we assume that rougher surface would result in a shallow interface states distribution as previously suggested, it is natural result that larger leakage current was easily observed with increasing applied voltage $V_{\max}$ and lower average field.

IV. CONCLUSIONS

The dc leakage current of Ta_2O_5 is related to the on the surface roughness and an enhanced roughness may results in a large amount of defects at and near surface. The increase of roughness is associated with the formation of wider interfacial region in spatial and broader distribution of interface trap depths. As a result, the slower transition slope in C–V (ac) and the lower phosphor turn-on field was resulted for TFEL devices under bipolar pulse driving. Finally, we observed the fact that these phenomena affect on average field within the phosphor through the change of polarization charge and leakage charge. Through these works, it was confirmed that interface roughness between phosphor and high dielectric constant insulating film (Ta_2O_5) is strongly related to surface or interface characteristics as well as the internal phosphor field for TFEL devices under short bipolar pulse driving. Therefore, we can suggest that internal characteristics can be controlled by interface roughness between phosphor and dielectric films and furthermore, the interface roughness of dielectric and phosphor layer is one of the physical factors to change the electrical performance of ACTFEL device.

Further study and experiments to clarify more details of the effect of the roughness on luminance characteristics are in progress.

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