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Transparent and Flexible Thin Film Electroluminescent Devices Using HiTUS Deposition and Laser Processing Fabrication

COSTAS TSAKONAS[1], STEVE WAKEHAM[2], WAYNE M. CRANTON[1,3], MIKE THWAITES[2], GABRIEL BOUTAUD[1], CARLY FARROW[1], DEMOSTHENES C. KOUTSOGEORGIS[1], AND ROBERT RANSON[1]

1 School of Science and Technology, Nottingham Trent University, Nottingham NG11 8NS, U.K.
2 Plasma Quest Ltd., Hook RG27 9UT, U.K.
3 Materials and Engineering Research Institute, Sheffield Hallam University, Sheffield S1 1WB, U.K.

CORRESPONDING AUTHOR: W. M. CRANTON (e-mail: w.cranton@shu.ac.uk)

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ABSTRACT Highly transparent thin film electroluminescent structures offering excellent switch on characteristics, high luminance and large break-down voltages have been deposited onto glass and flexible polymeric materials with no substrate heating using high target utilization sputtering. Deposition of ZnS:Mn as the active light emitting layer and Y_2O_3, Al_2O_3, Ta_2O_5, and HfO_2 as dielectric materials arranged in single and multiple layer configurations were investigated. Devices incorporating Al_2O_3, HfO_2 quadruple layers demonstrate the highest attainable luminance at low threshold voltage. Single pulse excimer laser irradiation of the phosphor layer prior to deposition of the top dielectric layer enhanced the luminance of the devices. The devices fabricated on glass and polymeric substrates exhibited a maximum luminance of 500 and 450 cdm^{-2} when driven at 270 V_{RMS} and 220 V_{RMS}, respectively, with a 1.0 kHz sine wave.

INDEX TERMS Electroluminescent device, thin films, laser processing, dielectric materials.

I. INTRODUCTION

Transparent displays are of interest for applications where subtle appearance and unobtrusive integration with the environment along with design aesthetics are requisites. Thin film electroluminescent (TFEL) transparent displays provide a method of applying a mature technology to this sector, and have recently been established as a commercial solution for niche markets where the environmental robustness of inorganic thin film electroluminescence is a desired attribute, such as in displays for use in space and deep ocean applications, arctic gas and oil fields, armored and excavation vehicles. The wide bandgap materials used for thin film electroluminescent devices make this technology particularly suitable for use in transparent configurations on glass substrates, where the electrodes utilized are transparent conducting oxides, and where the use of glass substrates facilitates high temperature processing to optimize the luminescent properties of the thin film phosphor. Application areas yet to be fully addressed by transparent displays are those that would benefit from flexible, conformable or rollable displays, which open up new functionality and design opportunities, as well as the potential for low production costs and reel-to-reel fabrication capability. The work presented here addresses this technological requirement via the demonstration of a low temperature method of fabricating a high luminance TFEL device on polymeric substrates via the use of deposition and processing techniques suitable for industrial production.

A standard TFEL device incorporating a double insulating layer structure consists of a wide band gap host doped with a luminescent center, sandwiched between two dielectric thin films. For applications that require high transparency

in the visible, ITO front and back contact electrodes are used. TFEL structures rely on the impact ionization of the phosphor by hot electrons that are emitted from defect states at the dielectric-phosphor interface. Given the optimum concentration and lattice position of the activator within the semiconductor, radiative emission of light will occur at a wavelength characteristic of the phosphor being excited. For the effective functioning of a TFEL device, it is essential that the dielectric layer is pinhole free, has a high dielectric constant and good breakdown field strength allowing for loss free acceleration of interface electrons to the energies required for phosphor excitation [1], [2].

Post deposition thermal annealing (at temperatures $\sim$ 500°C) can be used to improve EL emission by increasing the density of active luminescent centers [3], [4]. However, this also has the effect of reducing the profile of defect electron states at the phosphor-dielectric interface, which are the source of excitation electrons, hence resulting in shallow turn-on characteristics and decreased luminance [4], [5]. Additionally, high temperature annealing cannot be employed for applications that demand temperature sensitive, flexible substrate materials. One solution, we have demonstrated, is to laser anneal the phosphor layer prior to deposition of the top dielectric layer. Laser processing has been used very successfully in the past to greatly improve the luminance of EL devices deposited using rf magnetron sputtering [6] and is an area of intense research for the highly localized processing of thin films in our group [7], [8]. The work presented here investigates the application of this technique to low temperature processing suitable to realize transparent flexible EL devices.

II. EXPERIMENT

Full details of the High Target Utilization Sputtering system (HITUS) are given elsewhere [9]–[12]. The plasma generation mechanism in the HITUS system produces a high density magnetized plasma (10^{13} ions cm^{-3}) without the requirement of a target to strike or maintain the plasma. Ion energies are below the sputter threshold and therefore sputtering can only occur once a sufficient bias is applied to the target.

Independent control of the plasma density and the flux and energy of the sputtered species are achieved via close control of a combination of processing parameters; RF power, DC power and process pressure. In addition, the magnetized plasma interacts with the substrate coating process in an atypical and beneficial manner compared to conventional sputter processes. An optimal balance of plasma density and deposition rate may be set to deliver a low energy 'plasma assist' to the deposition process without the need for substrate bias. This beneficially impacts the thin film coating properties and enhances reactive deposition processes thereby resulting in the ability to coat fast deposition rate, high density films on to temperature sensitive polymeric substrates.

All the films grown in this work were deposited reactively from a metal alloy target in a gas environment consisting of argon and either oxygen or hydrogen sulphide (H_2S). An 800 nm ZnS:Mn film sandwiched between two dielectric layers, each 300 nm in thickness, has been used for all the devices investigated here. The dielectric layers were composed of either a single material or two materials arranged as a bilayer or twin bilayers in an attempt to reduce pinholing and maximize breakdown strength. The total dielectric thickness was maintained at 300 nm for all devices. ITO front and back contact electrodes were each 200 nm thick.

A TTi TG1010 programmable function generator and a voltage amplifier were used to drive the EL devices. A Minolta LS-110 luminance meter was used for recording the luminance of the samples. The devices were all connected in series to a current limiting 100 kΩ resistor. All EL measurements were carried out in a darkened room using a sinusoidal waveform with frequency 1.0 kHz and voltage increments of 2.85 V_{RMS} at 3 to 5 s duration.

The transparency of the films was measured using an Avaspec-2048 fiber optic spectrometer. All measurements were made with reference to the substrate and hence represent the transparency of the coating alone.

Laser processing was achieved using a 20 ns Lambda Physik LPX305i excimer laser (λ = 248 nm) in a pressurized chamber filled with Argon at 150 psi. XPS was performed using a Kratos Axis Ultra DLD with a monochromated aluminium x-ray source and delay line detection system. Cross-sectional TEM analysis was undertaken using a JEOL 2000FX operated at 200 keV.

III. RESULTS

A. ZNS:MN PHOSPHOR LAYER OPTIMIZATION

A.1. PL OPTIMIZATION

Single layers of ZnS:Mn were initially deposited onto glass microscope slides and optimized for photoluminescence (PL), with excitation via use of a cw-HeCd laser at 325 nm emission. The brightness of the PL emission was measured as a function of H_2S gas flow rate, Mn concentration in the sputter target, film thickness, and power applied to the target during growth. An improvement in optical transmission for higher H_2S flow rates is observed as the films make the transition from metallic Zn:Mn to stoichiometric ZnS:Mn. A corresponding increase in the PL intensity is also observed. For optimum PL, the H_2S flow rate was steadily increased until the films became fully transparent but not to the point that the target became poisoned.

A.2. DIELECTRIC OPTIMIZATION

Optimized efficiency of the EL devices requires that (a) the dielectric layer should be able to hold a charge density at least 3 times that of the phosphor layer and (b) the dielectric layer should be approximately 200 nm thick when the dielectric constant is about 10 [13]. Hence, there is a compromise between the thickness optimization of the phosphor layer for enhanced luminescence and the optimum for device stability and drive voltage. The primary disadvantage of increasing

the thickness of the ZnS:Mn is that this reduces the capacitance of the device and hence the threshold voltage for light emission will be higher. Conversely, for EL devices with a homogeneous phosphor layer, a thicker film will contain more luminescent centers that can contribute towards enhanced light emission.

Due to the importance of the phosphor-dielectric interface, several different dielectrics have been investigated in conjunction with the optimized ZnS:Mn. It is widely accepted that above the threshold voltage for EL, the field within the phosphor layer remains constant at approximately 1.5 $MVcm^{-1}$. This is known as the field clamping effect [13]. It is reported that field clamping effects are not observed if the density of interface states is too low or space charge formation is present in the phosphor layer, as with ALE deposited ZnS:Mn [14], [15]. The insulating layers must support any additional applied field and hence must exhibit high electric breakdown field strength. It is also important that they possess a high relative dielectric constant and are free from pinholes and contaminants that may cause device instability or breakdown. For applications requiring transparency on low temperature flexible substrates, additional constraints are that all of the device materials must be deposited with no, or limited, substrate heating and with high visible transparency. For this study, metallic targets were reactively sputtered in an $Ar:O_2$ plasma to produce the oxide films.

Dielectrics investigated were yttrium oxide (Y_2O_3), aluminum oxide (Al_2O_3), hafnium oxide (HfO_2) and tantalum oxide (Ta_2O_5). Aluminum oxide and hafnium oxide were singled out as having the highest break down fields, 2.1 $MVcm^{-1}$ and 3.5 $MVcm^{-1}$ respectively. The dielectric constants produced agree well with published data and they were $\varepsilon_r = 8 - 12$ [16], [17] for aluminum oxide and $\varepsilon_r = 20 - 30$ for hafnium oxide [9], [18]–[20]

B. ELECTROLUMINESCENT DEVICES

B.1. AS DEPOSITED DEVICES

To enable a direct comparison of the effect of the different dielectric materials devices were fabricated on glass using the four investigated dielectric materials. Each device was fabricated using the same ITO and ZnS:Mn processes. One way to reduce the threshold voltage is to use dielectric layers with high dielectric constant. As expected devices incorporating hafnia and tantala, having high dielectric constants, showed low threshold voltage while those with alumina, having the lowest dielectric constant, showed the highest threshold voltage. Another way to reduce the threshold voltage is clearly to reduce the thickness of the ZnS:Mn and dielectric layers. However, a reduction in phosphor thickness will result in a reduction in luminance and decreasing the thickness of the insulating layers can lead to instability as device breakdown is more likely to occur. Other factors that influence the threshold voltage relate to the interface states at the phosphor-dielectric layer boundary. If the interface states are shallow then a lower field will be required to cause electrons to overcome the energy barrier and travel through the phosphor layer. The gradient of the L-V curve for voltages immediately above threshold is dictated by the energy distribution of electrons in the defect states [21].

The spread of electron states in all of the devices presented here is significantly affected by the dielectric layer that is used, which will impact on the interface. Given that the drive conditions, layer thicknesses and phosphor properties are the same for all devices, the difference in attainable luminance is likely to originate from differences in electron states at the dielectric-phosphor interface [22]. For the devices fabricated using Hafnia a peak brightness of 86 cdm^{-2} is observed before the devices break down. This is compared to 75 cdm^{-2}, 49 cdm^{-2} and 24 cdm^{-2} for devices incorporating tantala, alumina and yttria respectively. The benefit of increased capacitance due to the materials with higher dielectric constants is evident in the concomitant reduction in drive voltages required. Alumina samples require a significantly higher drive voltage, due to the lower dielectric constant, but they also showed increased resilience against breakdown surviving voltages as high as 280 V_{RMS} Hence, it would be beneficial to combine materials for optimized performance and stability properties.

By incorporating alternating layers of Hafnia and Alumina into the dielectric stack, we can take advantage of both the high dielectric constant of the Hafnia and the high breakdown electric field strength of the Alumina. This was investigated using both single and double bilayers of alternating Hafnia and Alumina. The double bi-layer structure is clearly visible from the cross sectional TEM image in Fig. 7.

The results show that devices fabricated with multiple dielectric layers yield a far greater stability and resistance to breakdown. The EL characteristics are also more consistent from one device to the next, showing improved reliability and reproducibility. The devices with single bi-layer dielectric structures exhibit a maximum luminance of approximately 100 cdm^{-2}, a threshold voltage of 129 V_{RMS} and survive to in excess of 225 V_{RMS}. The devices with the double bi-layer dielectric structures exhibit a maximum luminance of approximately 100 cdm^{-2}, a threshold voltage of 138 V_{RMS} and survive to in excess of 233 V_{RMS}.

Fig. 1 shows the luminance vs applied bias for two optimized devices with single bi-layer (double) and twin bi-layer (quad) dielectric structures on glass substrates. It is possible to increase the maximum achievable luminance by increasing the frequency of the sine wave drive to 10 kHz. This results in the luminance of the twin bi-layer devices increasing to as much as 250 cdm^{-2}. When ITO is used for the front and rear contact electrodes, these devices have a transparency of approximately 90 % at 600 nm.

B.2. LASER ANNEALED DEVICES

Laser annealing of the upper surface of a ZnS:Mn thin film sandwiched between two dielectric layers has been shown to enhance the electroluminescent intensity of the completed

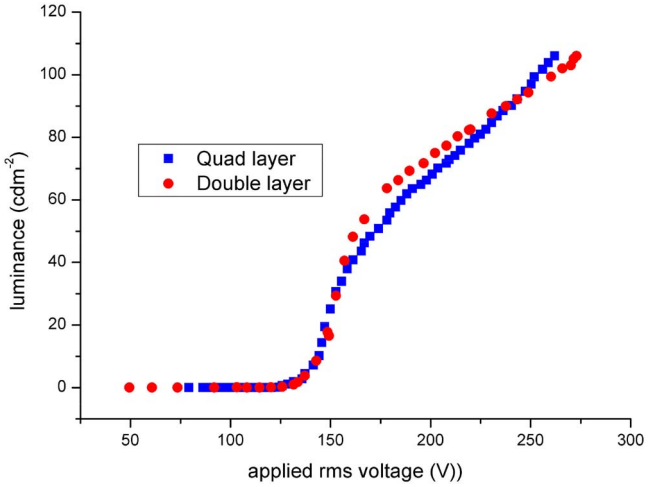

FIGURE 1. L-V characteristics for devices on glass substrates with double and quadruple dielectric layers of Hafnia/Alumina with an applied sinusoidal waveform of 1 kHz.

EL device by at least a factor of three [23], [24], when films deposited by RF magnetron sputtering were irradiated with single and multiple pulse irradiations at fluences of up to 1.2 Jcm^{-2} [25].

The electroluminescence response of a TFEL device, however, depends not only on optimization of the phosphor layer through crystallization but also on the energy configuration of the dielectric-phosphor interface states. Laser annealing has been observed previously to modify the ZnS crystal structure, in the top layers, from predominantly cubic in the as-deposit state to a more mixed phase cubic/hexagonal structure [26]. Since a 248 nm laser pulse has a considerably smaller penetration depth than the phosphor thickness, a process employing successive annealing of the phosphor layer as it is being deposited by RF magnetron sputtering was studied. To investigate this, the phosphor layer was deposited as four sub-layers (four separate depositions), each of 200 nm thickness, and laser irradiation of selected layers was undertaken prior to deposition of the next layer. Fig. 2 shows the photoluminescence of the 800 nm thick phosphor layer following this mixed processing and deposition approach. The results are from various combinations of laser annealed sections using different numbers of pulses and fluence levels. Fluence levels in the range 0.4 to 1.2 Jcm^{-2} and one, two and three pulses were used. Eight combinations of successive layer annealing were investigated. The particular layer combination is shown on the horizontal axis of Fig. 2 and is characterized by the letters A to H. The number indicates how many pulses were used. The fluence levels are plotted against the oblique axis while the vertical axis shows the photoluminescent intensity. The experiment produced a survey of 144 combinations of fluence, annealed layers and laser pulses. The results, in general, indicate that the photoluminescence depends on the total annealed thickness of the phosphor layer and is highest when medium fluences and three laser pulses are used with the highest PL being produced by the sample annealed at 0.8 Jcm^{-2} and 3 pulses in configuration D where the whole thickness of the sample was laser processed (i.e., sample D-3) as is expected. Furthermore these data show that annealing of the total thickness of the phosphor layer reduces the breakdown voltage of the device while the luminance stays at low levels. Hence, optimization of the photoluminescent intensity is not a required condition for the concurrent optimization of the electroluminescence.

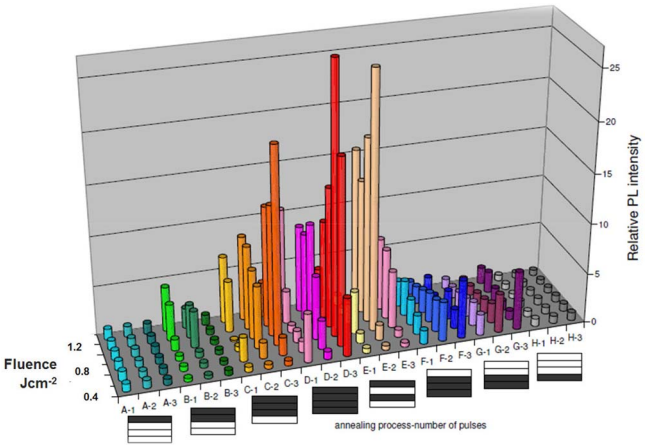

FIGURE 2. Photoluminescent intensity of the ZnS:Mn layer using various pulses (denoted by the numbers 1 to 3 on the x-axis) and laser fluences for different ZnS:Mn layer configurations, which are indicated by the letters. A-2 for example represents the sample with the top layer processed by 2 pulses.

Fig. 3 shows the electroluminescence of completed TFEL devices using the structures in Fig. 2 when three laser pulses have been used. For these devices, based on the PL results of Fig. 2, we might expect that the EL performance would follow a similar trend. However this is not the case as the EL performance is also affected by the electronic nature of the interfaces. Hence the results confirm the importance of the interface and indicate that when the dielectric layers either side of the phosphor layer are in contact with a laser annealed ZnS:Mn interface, the device survives only low fluences whilst the brightest devices have one of the interfaces formed from as-deposit material. Transient Electroluminescence analysis confirms this result showing very low EL originating from the laser annealed interface. Most of the EL is occurring when injection is taking place from the opposite interface indicating again the importance of the interface states position in the energy gap. Deep interface states result in highly energetic electrons and, laser annealed ZnS:Mn results in more efficient phosphor layer emitting high levels of EL compared to non-annealed and thicker layer.

The ZnS:Mn films were also investigated using surface XPS. The as-deposited films were sulphur rich with the relative sulphur content dropping as laser fluence increases (i.e., Zn/S varied from 0.36 - 0.46 with increasing laser fluence). Similar results were obtained for the RF sputtered films

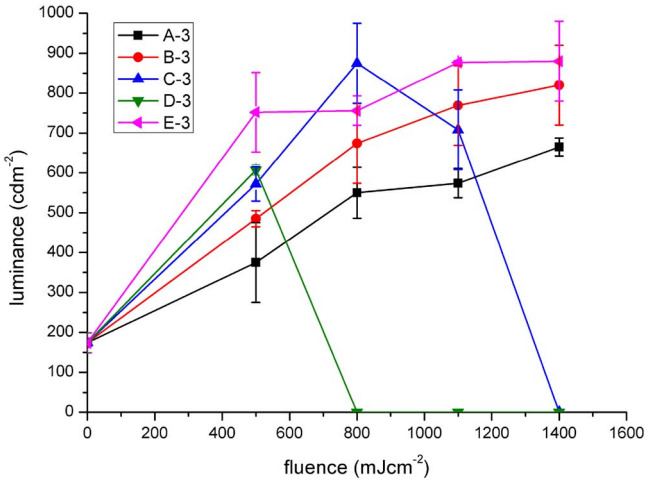

FIGURE 3. Electroluminescent intensity of TFEL devices processed by laser irradiation of selected ZnS:Mn sub-layers (as per Fig 2 labeling) using three laser pulses for each treated layer.

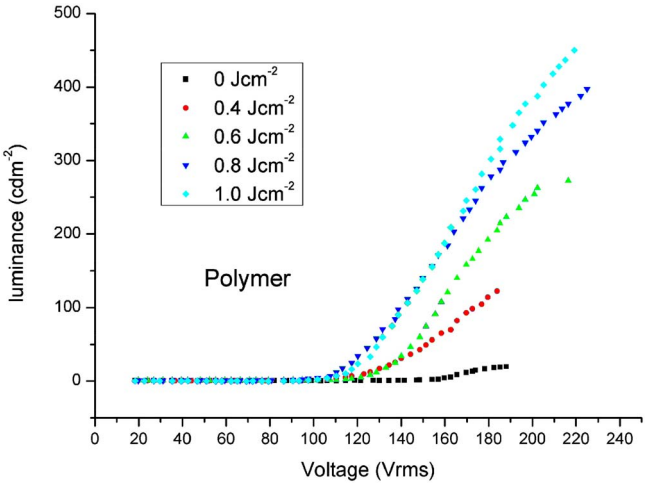

FIGURE 5. L-V characteristics for devices on the same polymeric substrate with quad dielectric layer annealed at various fluences using one pulse with an applied sinusoidal waveform of 1 kHz.

(i.e., sulphur content decreased with thermal annealing temperature). It is believed that the existence of Zn vacancies in S-rich films facilitates the diffusion of Mn atoms to substitute Zn atoms [27] in the crystal lattice and subsequently increase luminescence efficiency.

Fig. 4 shows the EL measurements of quad devices with hafnia and alumina dielectric layers on glass where the phosphor layer has been laser annealed at various fluences using one pulse. The increase of the maximum achievable luminance compared to the as deposit device is 18-24 times.

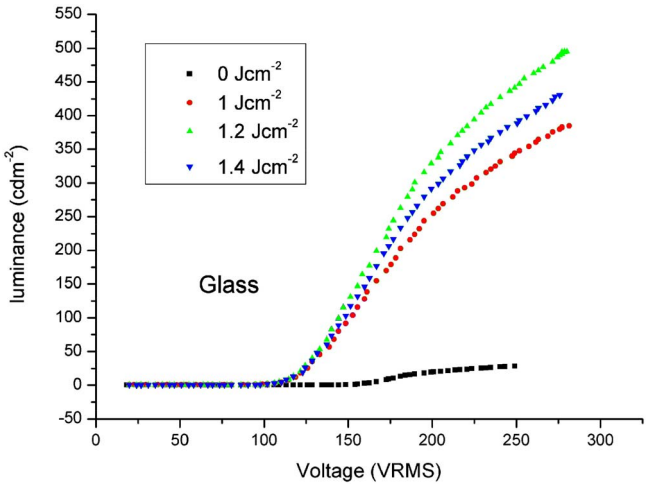

FIGURE 4. L-V characteristics for devices on the same glass substrate with quad dielectric layer annealed at various fluences using one pulse with an applied sinusoidal waveform of 1 kHz.

Fig. 5 shows the EL results of quad devices that were grown concurrently with the above devices on PEN substrates for direct comparison. Laser annealing of the phosphor layer again increases the luminance by similar factors.

To illustrate how the low temperature deposition and laser processing can be used to fabricate high luminance transparent EL devices on plastic substrates, Fig. 6 shows an image of a circular transparent TFEL device with diameter of 3 mm on a PEN substrate, showing the Mn electroluminescence.

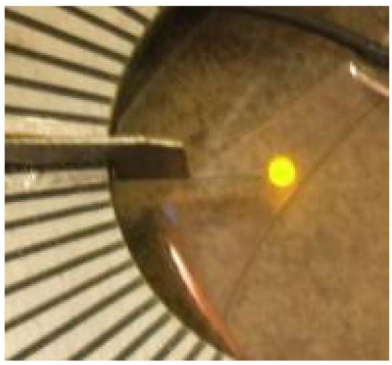

FIGURE 6. Example of transparent TFEL device with quad dielectric layers on a polymeric substrate and with the top ZnS:Mn layer annealed at 1.0 mJcm^{-2} and one pulse.

B.3. TEM RESULTS

Figs. 7 and 8 show cross sectional TEM images of three quad layer devices deposited onto the same PEN substrate: Fig. 7 showing the unannealed/as-deposited structure and Fig. 8 showing an area that has been laser processed at 0.4 Jcm^{-1}.

The thickness of the phosphor layer is about 715 nm. Tapered grain growth from fine grains at the bottom interface to larger grains at the top of the ZnS:Mn film is evident. For the laser processed regions, the grains in the modified zone have a much lower defect density than the underlying as-deposited layer and the upper surface exhibits reduced roughness. The modified zone extends to between

about 120 nm and 150 nm (for 0.4 Jcm^{-2} and 1.0 Jcm^{-2} respectively) following laser processing, which is consistent with the expected absorption of the 248 nm laser irradiation. Electron diffraction d-spacings calculations show that the crystal structure of both the modified and non-processed regions is hexagonal, in contrast to RF sputtered films that are mainly cubic in the as-deposit state [26].

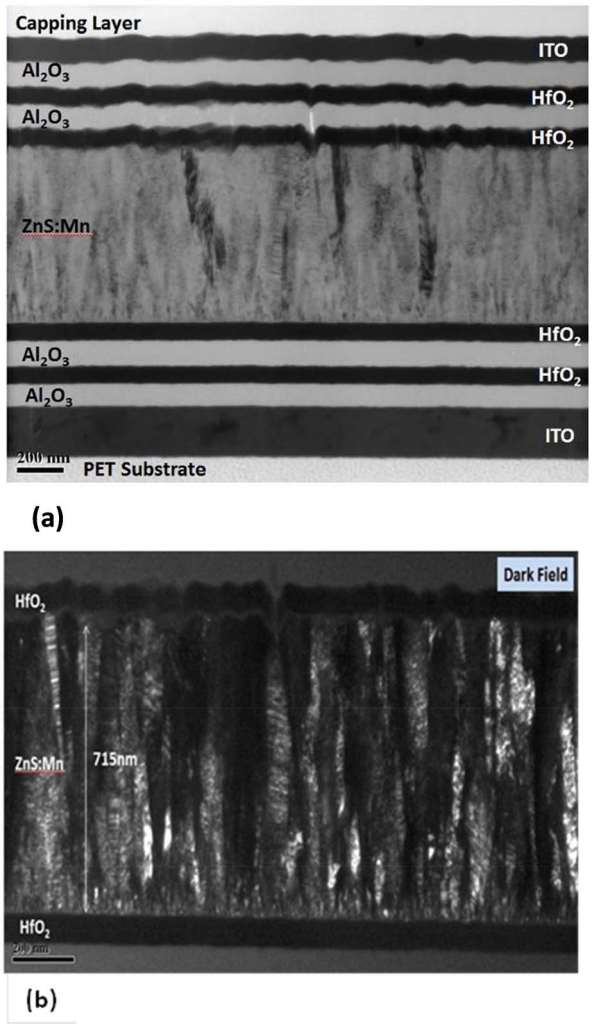

FIGURE 7. (a) TEM images of a non-processed TFEL device with quad layers on polymeric substrate, showing the stacked layer structure of the device, and (b), dark field image highlighting the course columnar grain structure of the ZnS:Mn thin film.

IV. CONCLUSION

TFEL devices have been fabricated on glass and planarised PEN substrate materials. A comprehensive study of the effect of using various dielectric materials in conjunction with a ZnS:Mn phosphor layer has been carried out. The characteristics of the EL devices are highly dependent upon the dielectric properties of the insulating layer used. The use of HfO_2 yields devices with the lowest threshold voltage (approximately 120 V_{RMS}) and a maximum luminance of 86 cdm^{-2}. When ITO is used for the front and rear contact

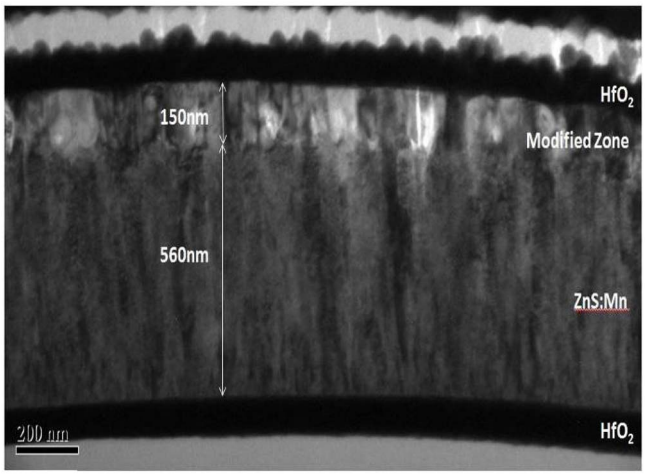

FIGURE 8. TEM image of a TFEL device with quad layers on polymeric substrate following laser processing at 1.0 Jcm^{-2} showing the clear delineation between the processed and unprocessed region of the ZnS:Mn, in this case extending to ~ 150 nm.

electrodes, the devices have a transparency of approximately 90 % at 600 nm. The low temperature process used throughout this work has enabled similar results to be achieved on glass and polymeric substrate materials demonstrating the capability of HiTUS to deposit transparent and flexible EL devices with good L-V characteristics.

Preliminary experiments have demonstrated a high luminance (~ 400 cdm^{-2} at 220 V_{RMS}) TFEL device on planarised PEN substrate achieved by laser annealing of the top 150 nm of the phosphor layer using 248 nm laser irradiation and one pulse, in ambient conditions. Transient luminance analysis showed that the origin of the electroluminescence was the top modified layers when electron injection is occurring from the bottom interface, due to deeper states originating at the bottom, unprocessed, interface.

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COSTAS TSAKONAS was born in Athens, Greece. He received the B.Sc. degree (Hons.) in electrical engineering from Patras University, Patras, Greece, and the Ph.D. degree in physical electronics from Bradford University, Bradford, U.K.

He worked in both industry and academia. He is currently a Research Fellow with Nottingham Trent University, Nottingham, U.K., where his research interests include the electro-optical characteristics of materials.

STEVE WAKEHAM received the master's degree and the Ph.D. degree in physics from the University of Reading.

He was a Post-Doctoral Researcher with the Multilayer Infrared Laboratory, University of Reading, before joining Plasma Quest Ltd., as a Senior Development Engineer in 2007. During his time at Plasma Quest Ltd., he has managed a successful government funded project on the deposition of metal oxides and sulphides for electroluminescent devices on plastic substrates and has been instrumental in the development of a diverse range of thin film materials using remote plasma. More recently, his internal research and development activities have led to a customer support roll for companies and universities throughout Europe and Asia.

WAYNE CRANTON was born in U.K. He received the Ph.D. degree from the University of Bradford, U.K., in 1995.

He is a Professor of Visual Technology with Sheffield Hallam University, and an Assistant Dean of Research and Business Development with the Faculty of Arts, Computing, Engineering and Sciences. He has worked in industry and academia in U.K., USA, and Canada. His research interests include the study of thin film materials for electronic displays, sensors, and light emitting devices, which has involved a number of collaborative applied research and development programmes on the deposition and processing of light emitting materials, dielectrics, and metal oxide semiconductors, with recent emphasis on the localized photonic processing of materials for low temperature fabrication of flexible electronics and displays.

MIKE THWAITES received the Ph.D. degree in physics from the University of Salford, in 1975.

He set up Plasma Quest Ltd., in 1998, after many years of experience in leading research laboratories. He developed HiTUS sputtering technology to overcome the numerous constraints of conventional magnetron sputtering. His expertise in plasma physics is valued by many universities and he holds Visiting Professorship with the University of Southampton, and Zhengzhou University, China, as well as an Honorary Professorship with Swansea University.

Prof. Thwaites is a fellow of the Institute of Physics and the Institute of Nanotechnology, a Chartered Physicist, a Chartered Scientist, and a member of the Institute of Electrical and Electronic Engineers.

GABRIEL BOUTOUD was born in France. He received the Ph.D. degree in growth optimisation and laser processing of thin film phosphors for electroluminescent displays from Nottingham Trent University, U.K., in 2010.

He was a Research Scientist Associate for a Knowledge Transfer Partnership Project between Essentra and Nottingham Trent University from 2010 to 2013, where he was responsible for developing expertise in light emitting pigments, devices, and associated materials used in the authentication of high value documents and products. He is currently a Research and Development Project Leader with Saint Gobain, U.K.

CARLY FARROW received the M.Eng. degree in system engineering from Loughborough University, U.K., in 2005, and the M.Sc. degree in engineering, cybernetics and communications and the Ph.D. degree in electroluminescent displays from Nottingham Trent University, U.K., in 2006 and 2014, respectively.

She is currently a Process Control Engineer with the Control and Instrumentation Engineering Department, Drax Power Ltd., Selby, U.K.

DEMOSTHENES C. KOUTSOGEORGIS was born in New York, NY, USA. He received the B.Sc. degree in physics from the University of Ioannina, Greece, in 1997, and the Ph.D. degree in materials science from Nottingham Trent University in 2003.

He has been a Lecturer with Nottingham Trent University since 2002, where he was promoted to a Senior Lecturer in 2005. In 2013, he became a Reader of Photonic Technologies with the School of Science and Technology, Nottingham Trent University. His research interests include fabrication and post deposition photonic processing of thin film materials for applications in plasmonics, electronics, optoelectronics, and sensors.

Dr. Koutsogeorgis is a member of SID, OSA, E-MRS, and MRS.

ROBERT RANSON received the B.Eng. degree (Hons.) in electrical and electronic engineering from the University of Bradford, in 1994, and the Ph.D. degree from Nottingham Trent University in 1999.

He has been a Senior Lecturer and a Principal Lecturer with Nottingham Trent University since 2000, where he is currently the Head of Computing and Technology, School of Science and Technology. His research interests include luminescent materials and engineering their properties for use within devices.

Dr. Ranson is a fellow of the Institution of Engineering and Technology, U.K.