Electrical and Luminescent Characteristics of a-SiC:H P-I-N Thin-Film LED's with Graded-Gap Junctions

Tean-Sen Jen, *Member, IEEE*, Nerng-Fu Shin, Li-Hong Laih, Yen-Ann Chen, Jyh-Wong Hong, and Chun-Yen Chang, *Fellow, IEEE*

***Abstract*—A-SiC:H p-i-n thin-film LED's (TFLED's) containing a single graded-gap p-i-n junction (SG) or double graded-gap p-i-n and i-n junctions (DG) have been postulated and fabricated successfully on indium-tin-oxide (ITO)-coated glass substrates, with a plasma-enhanced chemical vapor deposition (PECVD) system. Some important characteristics and related physics of these two types of TFLED's are presented and discussed. At an injection current density (J) of 600 mA/cm^2, the brightness (B) of the SG and DG TFLED's obtained were 30 and 207 cd/m^2, respectively. This significant improvement of brightness, as compared to those of the previously reported TFLED's with a highest brightness of 20 cd/m^2, could be ascribed to the reduced interface states with the graded-gap junctions, lower contact resistance between ITO and p-layer due to a plasma treatment of ITO prior to p-layer deposition, post metallization annealing of thermally evaporated Al on n-layer, and higher optical gaps ($E_{\rm opt}$'s) of the doped layers employed. The slopes of the nearly linear B–J relationships show the diode factor very close to unity for the fabricated SG and DG TFLED's. This implies that the electroluminescence (EL) mechanism of these TFLED's might be a tail-to-tail-state recombination. In addition, the conduction currents of these TFLED's are almost temperature dependent, and that of the DG TFLED might consist of an ohmic current and a space-charge-limited current (SCLC) within the lower and higher applied-bias regions, respectively.**

I. Introduction

With a lack of long-range ordering with structural symmetry, various multi-component amorphous semiconductors having tailor-made optoelectronic properties can be produced. Its deficiency of long-range periodicity also relaxes the k-selection rule for optical transition. This results in an increase of optical absorption coefficient, and could be used to yield a high luminescent efficiency [1].

Infrared electroluminescence (EL) in a Schottky-barrier interface of a-Si:H at low temperature was first reported in 1976 [2], and then also observed in an a-Si:H p-i-n junction [3]. After that, various visible a-SiC:H-based thin-film light-emitting diodes (TFLED's) were developed [4]–[10], including basic p-i-n [4], p-i-n embedded with hot-carrier tunneling injectors (HTI's) at p-i and/or i-n interfaces [5], p-i-n with microcrystalline- (μc-) p-a-SiC:H layer [6], μc-n-a-SiC:H layer [7], a-SiC:H/a-SiN:H luminescent multilayer [6], i-a-SiN:H luminescent layer [8], n-a-Si:H layer deposited with heavy H_2-dilution process [9], and μc-n-a-Si:H layer [10]. Among these TFLED's, the maximum achievable brightness (B) reported is around 20 cd/m^2 at an injection current density (J) of 1000 mA/cm^2 [5].

The EL of a forward-biased basic p-i-n a-SiC:H TFLED comes from radiative recombination of carriers in the luminescent i-layer. However, due to the limitation of valence-electron controllability, there is a notch barrier at the p-i and i-n junctions, respectively. These notch barriers retard the carrier transport and hence the EL intensity is weak [4]. Therefore, it is necessary to reduce notch barriers to enhance carrier transport and hence obtain a higher EL intensity at a lower forward-bias of TFLED. Consequently, a single graded-gap (SG) p-i interface can be used for improving the brightness of an a-SiC:H p-i-n TFLED [11], since the increased hole current plays an important role in radiative recombination and, hence, EL properties of TFLED's [5], [11]. In addition, a double graded-gap (DG) structure which has graded-gap p-i and i-n interfaces enhances the device brightness significantly and reduces its EL threshold voltage ($V_{\rm th}$) substantially [12]. As compared to the complex fabrication process for the a-SiC:H TFLED with barrier-layer structures [5], [9], the simple fabrication process for the graded-gap structures provide a higher benefit and reproducibility in production. The characteristics of these two kinds of a-SiC:H TFLED's, such as I–V and B–J relationships, EL spectra, current-conduction mechanism, and stability are presented in detail in this paper.

II. Device Fabrication

A schematic cross section of the DG TFLED is shown in Fig. 1 [12], whereas that for the SG TFLED has only a single graded-gap junction at the p-i interface and its i-layer thickness is 470 Å [11]. After a standard cleaning process, an indium-tin-oxide (ITO) coated glass substrate was transferred into a plasma-enhanced chemical vapor deposition (PECVD) system (ULVAC CPD-1108D) which was used for depositing various amorphous thin-films. The deposition conditions and optical gaps ($E_{\rm opt}$'s) of various layers for the SG and DG TFLED's were listed in Table I.

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T.-S. Jen, N.-F. Shin, L.-H. Laih, Y.-A. Chen, and J.-W. Hong are with the Department of Electrical Engineering, National Central University, Chungli 32054, Taiwan, R.O.C.

C.-Y. Chang is with the Institute of Electronics, National Chiao-Tung University, Hsinchu 30050, Taiwan, R.O.C.

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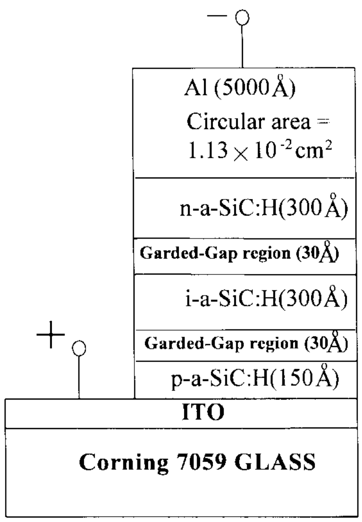

Fig. 1. The schematic cross section of the DG a-SiC:H p-i-n TFLED.

TABLE I
THE DEPOSITION CONDITIONS AND OPTICAL-GAPS OF VARIOUS LAYERS FOR THE SG AND DG TFLED's

Layer	Flow–rate ratio (sccm)	Pressure (torr)	G.R.* (Å/s)	E_{opt} (eV)	P_{rf}† (mW/cm²)
p(SG)	$SiH_4:C_2H_2:B_2H_6 = 200:6:38$	0.3	1.60	2.40	64
p(DG)	$SiH_4:C_2H_2:B_2H_6 = 200:6:72$	0.38	0.83	2.47	16
i(SG)	$SiH_4:C_2H_2 = 200:6$	0.3	1.25	2.50	64
i(DG1)	$SiH_4:C_2H_2 = 200:10$	0.3	0.52	2.61	16
i(DG2)	$SiH_4:C_2H_2 = 200:16$	0.3	0.65	2.65	16
n(SG)	$SiH_4:CH_4:PH_3 = 200:8:72$	1.0	1.40	2.00	112
n(DG)	$SiH_4:C_2H_2:PH_3 = 200:6:144$	0.46	0.45	2.40	16

*: Growth rate
†: RF power density

Substrate temperature: 180 °C
Process gases:
1: SiH_4 gas: $96\%H_2+4\%SiH_4$
2: B_2H_6 gas: $99\%H_2+1\%B_2H_6$
3: PH_3 gas: $99\%H_2+1\%PH_3$
4: C_2H_2 and CH_4 gases: pure

Before depositing the p-a-SiC:H layer of DG TFLED's, H_2-plasma bombarding to the ITO surface in the PECVD system was used for reducing the contact resistance between the ITO electrode and p-layer [12], [13]. It was found that the contact resistance between p-layer and ITO electrode was 4.42×10^6 Ω-cm without H_2-plasma treatment, and reduced to 7.72×10^5 Ω-cm with H_2-plasma treatment. This can be ascribed to a) the stabilization effect resulting from the reaction of hydrogen links with silicon dangling bonds on the surface being covered with hydrogen atoms, and b) the cleaning effect coming from the reactive hydrogen interacts with impurities on the surface, such as oxygen and carbon, and then removes them by cleaning volatile species [13]. The reason to replace the CF_4-O_2-plasma treatment, which was used in fabrication of the SG TFLED [11], by an H_2-plasma treatment is that it has less contamination to the following films to be deposited [13], [14].

To obtain the higher $E_{\rm opt}$'s of doped layers and hence lower the barrier height at p-i interface, the carbon source gas used was C_2H_2 rather than CH_4 [4], since the a-SiC:H film deposited with C_2H_2 has a small $E_{\rm opt}$-narrowing caused by boron doping and also a higher conductivity than that of the one deposited with CH_4 [15]. It is found that the luminescence intensity of the deposited film increases with decreasing RF power [16]. Therefore, the used RF power density was decreased from 64 mW/cm^2 for the SG TFLED to 16 mW/cm^2 for the DG TFLED's. The $E_{\rm opt}$'s, determined from individual Tauc's plots [17], of different layers for the SG and DG TFLED's are also summarized in Table I. As also observed in our laboratory, the i-a-SiC:H layer deposited with a lower RF power density tended to have a higher $E_{\rm opt}$ than that of the one deposited with a higher RF power density. Experimentally, the i-a-SiC:H layers deposited with 4% SiH_4 in H_2 (200 sccm) and pure C_2H_2 (6 sccm) had $E_{\rm opt}$'s of 2.50 and 2.57 eV for used RF power densities of 64 and 16 mW/cm^2, respectively.

After depositing all of the a-SiC:H layers, an Al film was thermally evaporated onto the n-layer, through a metal mask, to form contact electrodes. Finally, the device was annealed in an H_2 ambient with an ULVAC TA-7000 rapid thermal annealing (RTA) system to improve the contact between the Al electrode and n-layer [12].

The finished devices were then characterized and their performances were compared. The presented EL data in this paper were the best one among at least ten devices of the same structure, and the performance variation of the successful devices was within 10%.

III. DEVICE OPERATION

Fig. 2 shows schematic optical-gap diagrams under thermal equilibrium and forward-bias conditions for the SG and DG TFLED's. The optical-gap diagrams of the SG TFLED were similar to those of DG one, except that the $E_{\rm opt}$'s of employed a-SiC:H layers were different and there was a potential step at the i-n interface as indicated by the dash lines.

For a basic p-i-n a-SiC:H TFLED, when the used carbon-source gas is CH_4, the $E_{\rm opt}$'s of doped layers are limited to 2.0 eV due to valency-electron controllability, while the i-layer $E_{\rm opt}$ can be 2.5 eV or more [4]. Consequently, as above-mentioned, notch barriers exist at the p-i and i-n interfaces [5]. It has been reported by D. Kruangam *et al.* [4], [5] and also H. Mimura *et al.* [18] that the notch barrier ΔE_v at the p-i interface is about three times larger than that ΔE_c at the i-n interface. Those band discontinuities at the p-i and i-n hetero-interfaces in a-SiC:H p-i-n junction were determined by means of the internal photoemission from the structures of Au/i-a-SiC:H/p-a-SiC:H and Au/i-a-SiC:H/n-a-SiC:H. In their determination, the x-intercept in a plot of (photoelectric yield)$^{1/3}$ versus photon energy gives the threshold photon energy which is equal to the sum of the optical gap of the n- or p-layer and the valence-band's discontinuity ΔE_v. According, the estimated band discontinuities were $\Delta E_v = 0.075$ eV and $\Delta E_c = 0.125$ eV for the SG TFLED, and on the other hand, $\Delta E_v = 0.105$ eV and $\Delta E_c = 0.0525$ eV for DG1 TFLED,

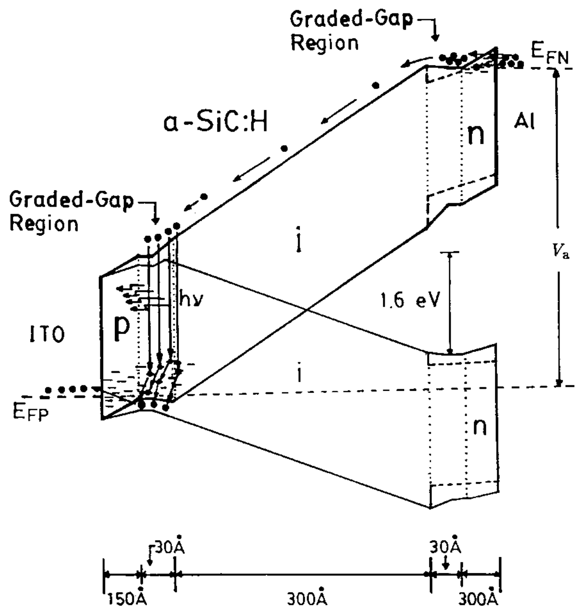

Fig. 2. The schematic optical-gap diagrams under thermal equilibrium and forward-bias conditions for the SG and DG a-SiC:H TFLED's.

and $\Delta E_v = 0.135$ eV and $\Delta E_c = 0.0625$ eV for DG2 TFLED, respectivity in this study. The EL comes mainly from the radiative recombination in the i-layer, especially near the p-i interface [5], since electrons have a higher mobility than that of holes [19]. To obtain a higher EL intensity, however, not only a lot of electrons but also many holes must be injected into the i-layer. As will be discussed in Section IV-B, the dominant carrier transport mechanism was single-carrier (electron) injection trap-free SCLC. The reduction of ΔE_c in DG TFLED's will increase the electron injection efficiency, hence enhance the EL intensity.

To reduce the effect of notch barrier on hole transport, a graded-gap p-i junction can be used [11]. In addition, an abrupt change of composition may induce many defects, and hence trapping centers at the interface. These trapping centers at the previously used step p-i interface, resulted from the interruption of RF power during the depositions of device's layers, degrade the EL intensity due to the increased nonradiative recombination. The graded-gap p-i junction obtained by a continuous deposition process can be employed to obtain a better interface property due to the reduction in effective interface recombinations.

As mentioned above the doped layers of DG TFLED's have higher $E_{\rm opt}$'s than that of the SG TFLED. Then, the efficiency of carrier injection from the doped layer into the i-layer could be improved with the lowering of effective energy-barrier at the interface of the doped layer and i-layer of the DG TFLED's. So, both of the a-SiC:H films deposited with a lower RF power density and a post-metallization annealing (PMA) process, which is intended to obtain a lower contact resistance between the Al and n-layer, benefit to obtain a higher EL intensity [12], [16]. It is also generally found, as the $E_{\rm opt}$ of a-SiC:H increases, the density of band tail states increases and its shape broadens [20]. Therefore, the utilization of an a-SiC:H i-layer with a higher $E_{\rm opt}$ (e.g., DG2 TFLED) can result in a higher EL intensity [12].

IV. Device Characteristics

A. J-V_a Characteristics and EL Intensities

The relationships of EL intensity $(I_{\rm EL})$ and injection current density (J) versus applied voltage (V_a) for the DG1, DG2, and SG TFLED's had been presented in [11] and [12]. For the DG1 TFLED, J increased rapidly when V_a exceeded 6.5 V, where $I_{\rm EL}$ began to increase. The J-V_a and $I_{\rm EL}$-V_a behaviors of the DG2 TFLED were similar to those of the DG1 TFLED. Since the i-layer $E_{\rm opt}$ of the DG2 TFLED was higher, its J was lower than that of the DG1 device under a certain V_a. The SG TFLED had a higher injection current density than those of the DG TFLED's, within the lower applied voltage region. This could be ascribed to its lower i-layer $E_{\rm opt}$, and hence lower resistivity, than those of the DG TFLED's. The EL threshold voltages V_{th}'s, where V_{th} was defined as the x-axis intercept of the straight portion in the $I_{\rm EL}$–V_a curve, were 11.2 V for the DG1 TFLED and 13.8 V for the DG2 TFLED, which were significantly lower than 32 V for the SG TFLED. This may be due to the incorporation of double graded-gap junctions, and the smaller i-layer thickness (300 Å) of the DG TFLED's. Also, the used step-gap i-n junction of the SG TFLED would degrade its EL property and result in an increase of its V_{th}.

We used an Oxford 1714 temperature-controlled liquid-N_2 crystat system to measure the device characteristics under various operating temperatures (T's). If the $\log(J/V_a^2)$ versus $(1/V_a)$ relationship of the SG TFLED at a certain temperature was checked by following the Fowler–Nordheim $(F$–$N)$ formula for about $V_a \geq 27$ V $(1/V_a \leq 3.6 \times 10^{-2}$ V$^{-1})$, then, as shown in Fig. 3, a fairly linear dependence was found [11]. However, this dependence on temperature implied its dominating electron current was not simply a F–N tunneling current, which is essentially temperature-independent [19], when V_a exceeded about 27 V. The injection current within the low bias region for the SG TFLED was essentially a thermionic emission current [11]. As shown in the inset of Fig. 3, $I_{\rm EL}$ of the SG a-SiC:H TFLED at a certain J increased with decreasing T.

Fig. 4 shows the J–V_a relationships of the DG1 TFLED, which were also temperature-dependent and similar to those of the SG TFLED. As obviously depicted in the inset of Fig. 4, the $I_{\rm EL}$ of DG1 TFLED at a certain J also increased with decreasing T. This phenomenon implied the radiative recombination efficiency at a lower temperature was higher.

B. Carrier Transport Mechanism

The conduction currents of the SG and DG TFLED's were temperature-dependent as mentioned above, so their conduction current possibly includes other current components or is dominated by other current transport mechanisms. In this section, only the J-V_a characteristics of the brighter DG TFLED was investigated from the point view of the space-charge-limited current (SCLC).

Referring to the schematic optical-gap diagram of the DG TFLED as shown in Fig. 2 under thermal equilibrium, there was a 1.6 eV difference in the electrostatic potentials of the n- and p-regions [12]. The injection current of the DG1 TFLED

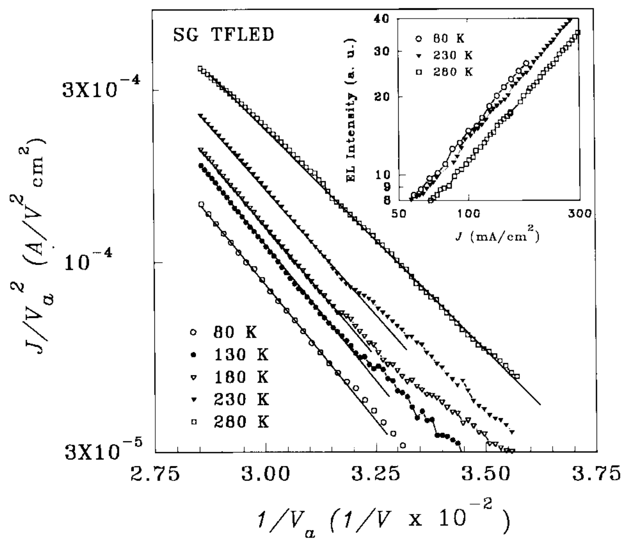

Fig. 3. The linear relationship of $\log(J/V_a^2)$ versus $1/V_a$ within the higher V_a region for SG TFLED under various temperatures. The inset shows the characteristics of EL intensity versus injection current density for the SG TFLED at various temperatures.

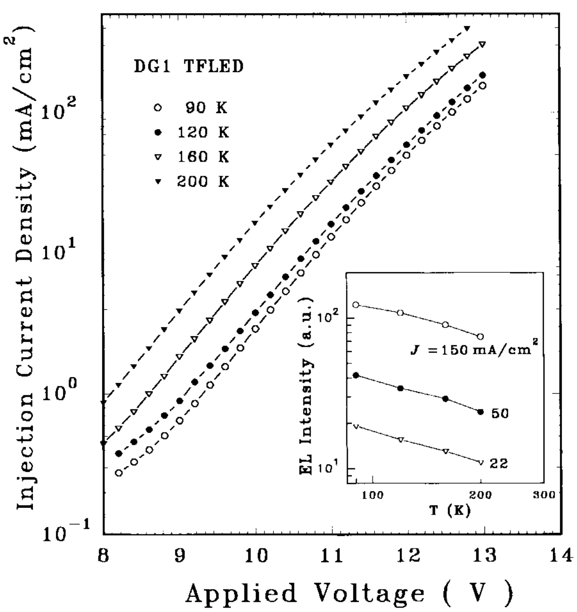

Fig. 4. The relationships of injection current density versus applied voltage for the DG1 TFLED at various temperatures. The inset shows the relationships of EL intensity versus temperature for the DG1 TFLED at various injection current densities.

increased very rapidly when V_a increased from 0 to 1.6 V. At $V_a = 1.6$ V, the flat-band condition was achieved. For 1.6 V $< V_a <$ 8 V, the injection current could become a SCLC with traps [17], [21], [22], and the Ohm's law would be observed because of the presence of thermally generated free carriers with a concentration n_o in the amorphous material [21], [22]. At $V_a = 8$ V, the trap-filled limit of amorphous semiconductor was reached. As $V_a > 8$ V, the average injected excess free-carrier concentration n_i could become comparable to n_o, a dominant single-carrier (electron) injection trap-free

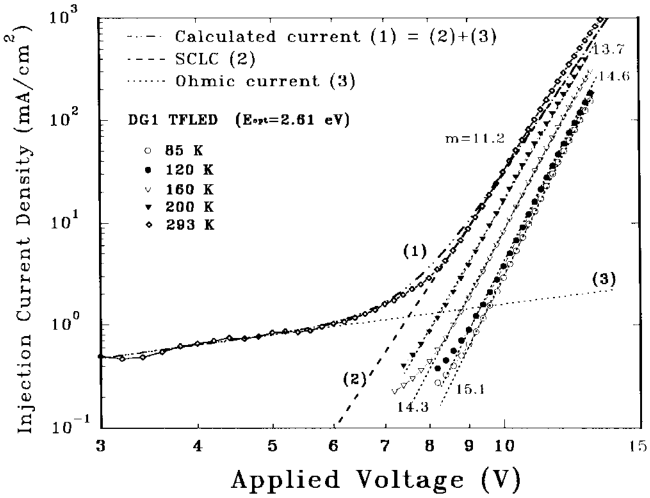

Fig. 5. The log-log plots of J–V_a curves for DG1 TFLED at various temperatures.

SCLC would be onset, so that the current rose rapidly [17], [21], [22], and the EL intensity started to increase.

Fig. 5 illustrates the calculated and experimental $\log(J)$ versus $\log(V_a)$ curves of the DG1 TFLED at various temperatures. At $T = 293$ K, a linear relationship of the J–V_a curve, which indicated an ohmic current, was observed at the lower voltage region from 3 to 6 V. Also, for all curves, an indication of SCLC by the power-law relationship of $J\alpha V_a^m$ was obtained in the higher voltage region. Here, $m[\equiv (l+1) > 2]$ value could be determined from the slope of the linear portion of curve (2) in Fig. 5.

As could be estimated from $l \equiv T_c/T$ [22] and $m = 11.2$ at $T = 293$ K, shown in Fig. 5, the characteristic temperature T_c equaled 2989 K. This T_c value, which reveals the trap-state distribution in the a-SiC:H material, was much higher than 1060 K obtained from i-a-Si:H. That meant the a-SiC:H film had a higher trap-state density within the gap than that of the a-Si:H film [22]. This would be caused by the incorporation of carbon atoms into the a-Si:H network [20]. Therefore, we concluded that the SCLC fitted very well to the current conduction of the DG TFLED within the higher applied-voltage region. However, as could be estimated from m's of Fig. 5, the T_c increased from 1199 to 1989 K as T increased from 85 to 293 K. This may be due to that the frozen carriers at low device operation temperatures would reduce the effective trap density within the gap and hence T_c.

C. EL Spectra

The EL spectra for the SG, DG1, and DG2 TFLED's can be found in [12] and [13]. The peak wavelength of EL spectrum of the SG TFLED (with an i-layer $E_{\rm opt}$ of 2.50 eV) located at 710 nm, which was in the red-light region. Its full width at half maximum (FWHM) was 230 nm. A red-orange color was observed by naked eyes, experimentally.

For the DG1 TFLED (with an i-layer $E_{\rm opt}$ of 2.61 eV), the EL spectrum peaked at 680 nm with a FWHM of 215 nm and revealed an orange color. On the other hand, the EL spectrum of the orange DG2 TFLED (with an i-layer E_{opt} of 2.65 eV) peaked at a longer wavelength (700 nm) and had a broader emission band (FWHM $= 240$ nm) than that of the DG1 TFLED. Such broad spectra of light emission could be primarily due to the wider energy-range of localized states in the i-a-SiC:H which had a higher carbon content and hence a higher E_{opt}. Also, the broader FWHM of the SG TFLED than that of the DG1 TFLED could be ascribed to the higher RF power density in thin-film depositions which increasing the defect density [15].

The EL spectra of these TFLED's peaked at around 1.7 eV, which was much lower than that of the E_{opt} of luminescent i-layer. This could be due to that the radiative recombinations were via the broad localized states in the amorphous i- and/or p-layers.

D. Stability

It was found that the EL intensity of DG1 decreased gradually within the first 1 min. from the beginning of operation. Then, its EL intensity becomes stable and almost keeps at a steady-state value of about 93% of the initial value when the driving time exceeding 5 min.

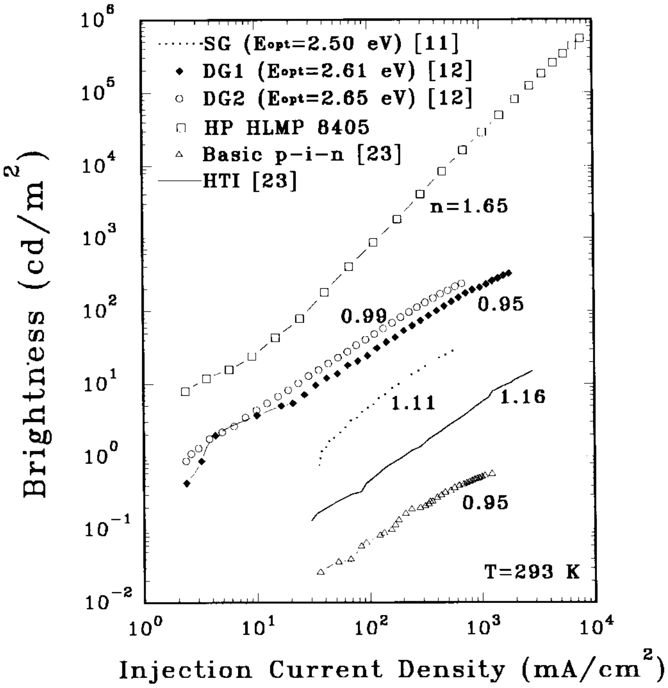

Fig. 6. The brightness comparison of the a-SiC:H basic p-i-n, HTI, SG, DG1, DG2, TFLED's, and HP HLMP-8405 orange LED.

V. Brightness Comparison

Fig. 6 shows the brightness (B) comparison of a-SiC:H basic p-i-n, HTI [23], SG, DG1, DG2, TFLED [11], [12], and packaged HP HLMP 8405 ultra-bright DH AS (double heterostructure on an absorbing substrate) AlGaAs orange LED. The DG TFLED's had much higher B than that of the SG TFLED, which was primarily due to the improvements of injection current and radiative recombination, as mentioned in Section III. For the DG1 TFLED, the B was 140 cd/m^2 at $J = 600$ mA/cm^2. The DG2 TFLED revealed a B of 207 cd/m^2 at the same J. The higher brightness of the DG2 TFLED as compared to that of the DG1 TFLED could be due to the wider energy-range with more localized states in the i-layer which increased the recombination probability. The B-J relationship of an a-SiC:H TFLED can be expressed as: $B \alpha J^n$, where the exponent n is 1 for a monomolecular recombination, 2 for a bimolecular recombination, and $0 < n < 2$ for a tail-to-tail-state recombination [4]. From Fig. 6, the estimated n's were very close to 1 for these a-SiC:H TFLED's, while the n value of the HP LED was 1.65. This revealed that the EL mechanism of the a-SiC:H TFLED's was due to the tail-to-tail-state recombination [4]. But, the EL mechanism of the HP LED was more closer to the bimolecular recombination.

VI. Conclusion

We have greatly improved the EL intensity of a-SiC:H p-i-n TFLED's by using the graded-gap junction. For instance, the brightness for the SG TFLED was 30 cd/m^2 at $J = 600$ mA/cm^2, which was more than two orders of magnitude higher than that of a basic p-i-n TFLED at the same injection current density. The highest brightness obtainable was 140 cd/m^2 for the DG1 TFLED and 207 cd/m^2 for the DG2 TFLED at the same injection current density. Further, the EL V_{th}'s of a-SiC:H TFLED's were significantly lowered by the adoption of graded-gap junctions. We ascribed the significant improvement of brightness and lowering of EL threshold voltage to the enhancement of carrier injection efficiency by introducing the graded-gap structures, the reduction of contact resistances by using in-situ H_2-plasma-treated ITO and an annealing process, the higher E_{opt} of doped layers which lowered the effective barrier height at interfaces, and the reduced defect density by decreasing the RF power density during thin-film depositions.

The current conduction mechanism of DG TFLED's had been briefly studied: the ohmic conduction dominated the carrier transport within the lower applied-voltage region, while the space-charge-limited current was the dominant current within the higher applied-voltage region where EL was observed. The estimated characteristic temperature T_c of an a-SiC:H equaled 2989 K at room temperature, which was much higher than that of undoped a-Si:H and showed a higher trap-state density within the gap of i-a-SiC:H films. The n's of fabricated TFLED's were very close to 1, which revealed that the tail-to-tail-state recombination was dominant.

The simple fabrication process employed and the improved EL properties of the DG TFLED's revealed would enhance the application potential of a-SiC:H p-i-n TFLED's significantly.

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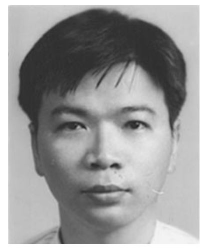
Tean-Sen Jen (S'90–M'95) was born in Chiayi, Taiwan, R.O.C., on September 14, 1966. He received the B.S. degree in electrical engineering and the Ph.D. degree in computer science and electronic engineering, from National Central University, Chungli, Taiwan, in 1988 and 1994, respectively. His interests include amorphous materials and devices and porous silicon.

In 1997, he joined Nan-Ya Technology Corporation, Taipei, Taiwan.

Dr. Jen is a member of Phi Tau Phi and the Chinese Institute of Electrical Engineers.

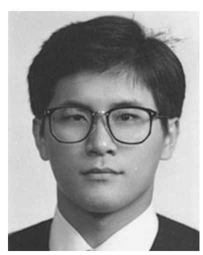


Nerng-Fu Shin was born in Tainan, Taiwan, R.O.C., on December 25, 1964. He received the B.S., M. S., and Ph.D. degrees from National Central University, Chungli, Taiwan, in 1987, 1989, and 1994, respectively.

In 1996, he joined Episil Technologies, Inc., Hsinchu, Taiwan. His present research topics include development of 20-GHz BJT IC's, and transport mechanism study of amorphous devices.

Dr. Shin is a member of Phi Tau Phi and the Optical Society of America.

Li-Hong Laih was born in Yurn-Lin Hsien, Taiwan, R.O.C., on September 17, 1970. He received the B.S. degree in electrical engineering from Chung-Yuan Christian University, Chungli, Taiwan, in 1993, and the M.S. degree in electrical engineering from National Central University, Chungli, Taiwan, in 1995, He is currently pursuing the Ph.D. degree at the Institute of Electrical Engineering, National Central University. His specialty is in the areas of amorphous materials, amorphous metal-semiconductor-metal (MSM) photodetectors, and porous silicon devices.



Yen-Ann Chen was born in Taiwan, R.O.C., on August 11, 1970. He received the B.S. and M.S. degrees in electrical engineering from National Central University, Chungli, Taiwan, in 1992 and 1994, respectively, and is currently pursuing the Ph.D. degree at the Institute of Electrical Engineering, National Central University. His specialty is in the area of amorphous materials and devices.

Jyh-Wong Hong was born in Taiwan, R.O.C., on May 24, 1947. He received the B.S. and M.S. degrees in electronics from National Chiao-Tung University, Hsinchu, Taiwan, in 1969 and 1972, respectively, and the Ph.D. degree from the Institute of Electrical Engineering, National Chen Kung University, Tainan, Taiwan, in 1990.

He joined Telecommunication Laboratories of the Ministry of Communications, in 1975, where he was engaged in the circuit design, fabrication of the semiconductor devices and development of hybrid integrated circuits. He joined the Electrical Engineering Department, National Central University, Chungli, Taiwan, as an Associate Professor in September 1985, and became a Professor there in 1992. His current research interests focus on amorphous materials and devices, and porous silicon.

Chun-Yen Chang (S'69–M'70–SM'81–F'88) was born in Kaoshing, Taiwan, R.O.C., on October 12, 1937. He received the B.S. degree in electrical engineering from National Chen Kung University, Tainan, Taiwan, in 1960, and the M.S. and Ph.D. degrees from National Chiao-Tung University, Hsinchu, Taiwan, in 1962 and 1970, respectively.

From 1962 to 1970, he was a Research Assistant, and then an Instructor, at Chiao-Tung University, working on organizing a semiconductor research laboratory. From 1966 to 1976, he was first an Associate Professor, and later a Professor, in solid-state electronics and semiconductor physics and technologies; he was also Chairman of the Department of Electrophysics, Chiao-Tung University, and has been a Professor and Director of the Institute of Electrical Engineering, National Chen Kung University, from 1977 to 1987, where he has established a strong research and development base in electrical and computer engineering. In 1981, he became a member of the technical staff at Bell Laboratories, Murray Hill, NJ, working in the VLSI Device Group. He was a Visiting Professor at the University of Florida, Gainesville, in 1987, and the University of Stuttgart, Stuttgart, Germany, in 1989, where he taught MBE technologies and devices. He has consulted ERSO/ITRI Nippon Seiki Co., UMC, and others on VLSI physics and technologies, including III–V compound devices and materials using MBE and MOCVD, and amorphous-silicon devices. He has taught the majority of senior semiconductor engineers in Taiwan since 1962, and has published more than 200 technical papers in international journals. He has contributed carrier transport theory and specific contact resistivity in metal-semiconductor systems. He invented a method for MOS surface stabilization, the method of low-pressure MOCVD using TEG source, amorphous Si phototransistors, and the bipolar-unipolar transistion negative resistance transistor (BUNDR). Currently, he is a Professor, the first Dean of the Collgee of Electrical Engineering and Computer Sciences, National Chiao-Tung University, where 172 faculties are involved, and the Director of National Nano Device Laboratory, Taiwan, R.O.C.

Dr. Chang is a member of Phi Tau Phi, the American Electro-Magnetics Academy, the Chinese Institute of Electrical Engineers, the American Physical Society, and the Electrochemical Society. He was elected an IEEE Fellow for "his contribution to semiconductor devices development and to education." He was the recipient of an Academic Achievement Award in engineering from the Ministry of Education, the Distinguished Research Award of the National Science Council, R.O.C., and the 1989–1990 International Traveling Award granted by the China Foundation to the distinguished scholars in the Republic of China.