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Efficient 1.54 μm light emitting diode with nanometer thick polycrystalline Si anode and organic sandwich structure

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This letter reports that the 1.54 μm electroluminescence efficiency of the organic light emitting diode (OLED) with a structure of nanometer thick polycrystalline silicon (NTPS)/NPB/ErQ/AlQ/Al is two orders of magnitude higher than that of the OLED with a structure of thick crystalline silicon/NPB/ErQ/Al, which is similar to the OLED reported in literature [Curry *et al.*, Appl. Phys. Lett. **77**, 2271 (2000)]. Such an improvement is mainly attributed to the fact that hole injection is controlled by NTPS anode and holes are blocked by AlQ to match electron injection, and a higher light out coupling as well. © *2006 American Institute of Physics*.
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In recent years, a monolithic integration of optics and electronics in a single Si chip has been pursued. However, Si is a poor light emitter due to its indirect band structure. Many approaches have been suggested to gain light from Si.[1–3] 1.54 μm Si-based light emission is of particular interest and realized by cooperation of organic electroluminescence (EL) technology with Si technology recently, where the emitters are erbium complexes.[4–6] Curry *et al.* have demonstrated an effective 1.54 μm organic light emitting diode (OLED) with a structure of crystalline Si (c-Si)/N,N'-diphenyl-N,N'-bis(3-methyl)-1,1′-biphenyl-4,4′-diamine (TPD)/tis(8-hydroxyquinoline) erbium (ErQ)/Al.[6] However, both the device current and voltage are high, resulting in a poor EL efficiency.

In this letter, we investigated how the following three factors impact the I-V characteristic and EL efficiencies of the 1.54 μm Si-based OLED: (1) electrical resistivity of the p-type c-Si anode, (2) replacing the c-Si anode with a nanometer thick polycrystalline Si (NTPS) anode, and (3) replacing organic double layers of N,N'-bis-(1-naphthl)-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB)/ErQ with triple layers of NPB/ErQ/tris(8-hydroxyquinoline) aluminum (AlQ).

The NTPS film was fabricated with the magnetron sputtering technology followed by the nickel-induced-crystallization process as follow. In a chamber with a base pressure of 1×10^{-8} Torr, 20 nm SiO_2, 75 nm Si, and 2 nm nickel layers were successively sputter deposited on the K9 glass substrates. The SiO_2 is to block the diffusion of impurities of glass into the Si layer. The sputtered Si layer has the same boron concentration as the 0.001 Ω cm boron doped Si target.[7] Then the samples were annealed at 500 °C in nitrogen atmosphere to form NTPS. We had no attempt to remove the native oxide on the NTPS surface, just as reported in Refs. 8 and 9. The sheet resistance of NTPS was measured to be $\sim$250 $\Omega/\square$ and the hole mobility was $\sim$3 cm^2/V s. The c-Si anodes were prepared as reported previously,[10,11] and Al contacts were formed on their backsides each with a window for light collection. The organic materials and the metals were thermal evaporated in a chamber with a base pressure of 5×10^{-6} Torr, and the typical deposition rate was 1–2 Å/s, monitored by a quartz crystal oscillator.

ErQ was synthesized by mixing erbium (III) chloride in 80% ethanol and 20% water solution with 8-hydroxyquinoline in ethanol. The infrared luminescence was measured by a liquid-nitrogen cooled Ge detector at room temperature.

In the following, three types of devices will be studied. Type A: 500 μm c-Si/60 nm NPB/60 nm ErQ/50 nm Al, in which the c-Si anode has an electrical resistivity of 0.01, 1, or 40 Ω cm, and so the resulting devices are, respectively, referred to as 0.01, 1, and 40 Ωcm device. Type A devices are similar in structure to the OLED ($\sim$0.01 Ω cm c-Si/40 nm TPD/50 nm ErQ/Al) reported in Ref. 6. Type B: 75 nm NTPS/60 nm NPB/60 nm ErQ/50 nm Al, i.e., the OLED having a NTPS anode and two organic layers. It is referred to as the NTPS-2 device. Type C: 75 nm NTPS/60 nm NPB/10 nm ErQ/50 nm AlQ/50 nm Al, i.e., the OLED having a NTPS anode and three organic layers. It is referred to as the NTPS-3 device. The c-Si anode or the NTPS anode is transparent for 1.54 μm light. The NPB layer is the hole-transport layer and the Al layer is the cathode in all the three types of devices. The 60 nm ErQ layer is used as the emissive and electron-transport layer in types A and B devices. A 10 nm ErQ layer served as the emissive layer and a 50 nm AlQ layer as the electron-transport layer in type C device.

Figure 1(a) depicts the EL spectra for the 0.01, 1, and 40 Ω cm c-Si devices, and Fig. 1(b) for the 40 Ω cm c-Si, NTPS-2, and NTPS-3 devices at the same current density of 2 mA/mm^2. In all the EL spectra, the peaks center at 1.54 μm, which is the characteristic emission of Er ions. It is

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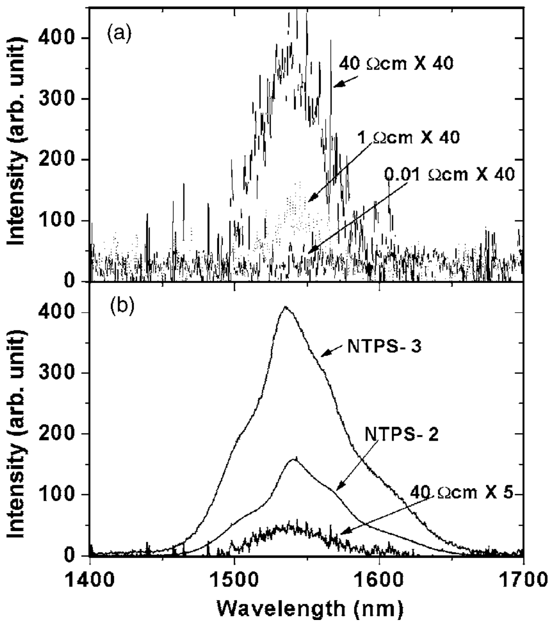

FIG. 1. (a) The EL spectra for the 0.01, 1, and 40 Ω cm devices at the same current density of 2 mA/mm^2. (b) The EL spectra for the 40 Ω cm, NTPS-2, and NTPS-3 devices at the same current density of 2 mA/mm^2.

generally accepted that this luminescence originates from a mechanism called the antenna effect.[12] Intersystem crossing occurs from the excited singlet formed by recombination of holes and electrons to a triplet state on the ligands, and then the energy of the triple state is transferred efficiently to the excited 4*f* states of the Er ions via a Dexter transfer. Finally, the intratransitions $^4I_{13/2}$-$^4I_{15/2}$ within Er ions produce the 1.54 μm luminescence.[12–14]

The current density, the infrared EL intensity, the current efficiency, and the power efficiency versus applied voltage are shown in Figs. 2(a)–2(d), respectively, for all the devices. It can be found that for the *c*-Si devices, the current density decreases with the increase of the electrical resistivity of *c*-Si anode, because the hole injection from the *c*-Si anode is reduced with increasing resistivity.[10] Moreover, the EL efficiency increases with increasing electrical resistivity of the *c*-Si anode, because the balance between electron and hole injections is improved by reducing hole injection. As a result, the 40 Ω cm device gives the best EL current and power efficiencies compared to the 0.01 and 1 Ω cm devices.

The infrared EL performance can be improved by using the 75 nm NTPS anode (NTPS-2 device) to replace the *c*-Si anode (40 Ω cm *c*-Si device) as shown in Figs. 2(a)–2(d). When the applied voltage is below 8 V, the current density of the NTPS-2 device is close to that of the 40 Ω cm device. When the applied voltage is between 8 and 15 V, a negative resistance occurs, the reason of which is not clear now and will be discussed elsewhere. When the applied voltage is above 15 V, the current density of the NTPS-2 device is markedly lower than that of the 40 Ω cm device. The maximum current and power efficiencies of the NTPS-2 device are 22 and 18 times, respectively, higher than that of the 40 Ω cm device with a *c*-Si anode.

One reason for the significant EL enhancement is that the balance between electron and hole currents in the OLED is improved. It is due to a low hole injection ability of NTPS, caused by the small carriers mobility. To show the hole injection ability of NTPS, we fabricated "hole-only" devices

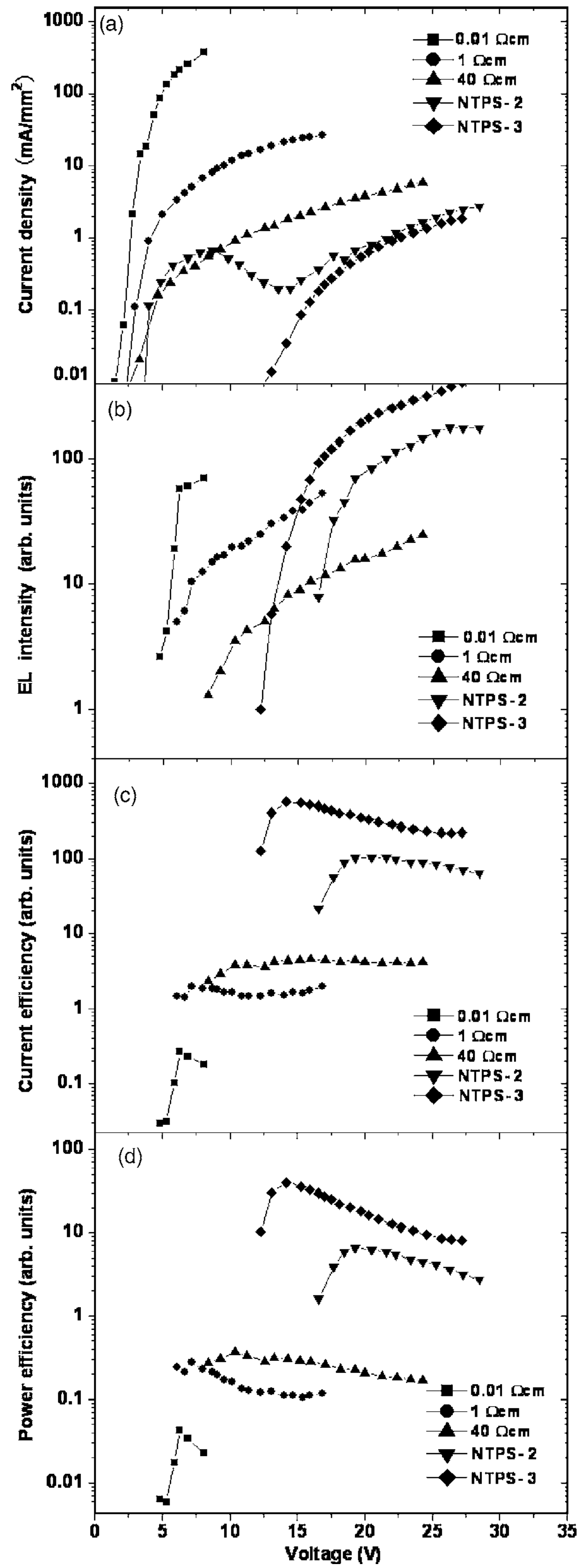

FIG. 2. (a) The current density, (b) the infrared EL intensity, (c) the current efficiency, and (d) the power efficiency vs applied voltage for 0.01 Ω cm, 1 Ω cm, 40 Ω cm, NTPS-2, and NTPS-3 devices.

with the NTPS or the *c*-Si anodes by replacing the Al cathode with a much higher work function metal Au.[15] As shown in Fig. 3, the hole current of device with NTPS anode is much smaller than those of any devices with *c*-Si anodes, that is, the hole injection ability of NTPS is much smaller than that of *c*-Si.

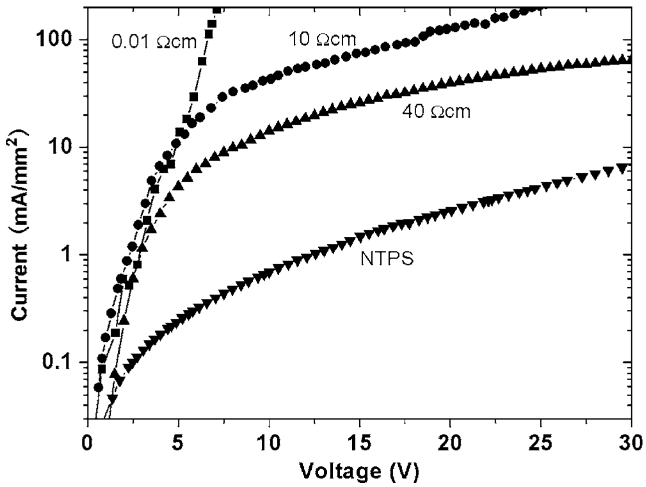

FIG. 3. The *I-V* characteristics of "hole-only" devices each with a NTPS or *c*-Si anode. (▼) 75 nm NTPS/60 nm NPB/60 nm ErQ/50 nm Au, (▲) *c*-Si (40 Ω cm)/60 nm NPB/60 nm ErQ/50 nm Au, (●) *c*-Si (1 Ω cm)/60 nm NPB/60 nm ErQ/50 nm Au, and (■) *c*-Si (0.01 Ω cm)/60 nm NPB/60 nm ErQ/50 nm Au.

Another reason for the significant EL enhancement is that the infrared light outcoupling efficiency has been improved markedly. We have calculated their light out coupling efficiencies at 1.54 μm using a classical technique.[16] The out coupling of the NTPS-2 device is larger than that of the 40 Ω cm device by 2.2 times.

The infrared EL efficiency of device can be improved further by improving the structure of organic layers. Figures 2(a)–2(d) demonstrate that, compared to NTPS-2 device, the current density of NTPS-3 device slightly decrease, but the EL intensity increases markedly at an applied voltage above 15 V, and the maximum current efficiency and power efficiency increase by six and five folds, respectively. It is because the AlQ layer is a better hole blocking layer than the ErQ layer,[5] which causes hole accumulation at the interface of ErQ/AlQ and enhances electron injection and EL intensity. Besides, the leakage hole current is reduced in NTPS-3 device, resulting in higher current and power efficiencies. Substituting an AlQ layer for an ErQ layer as the electron-transport layer affects hardly the electron injection into the emissive layer, because lowest unoccupied molecular orbital (LUMO) and electronic structure of ErQ are similar to those of AlQ.[17] As a result, the emissive layer ErQ can efficiently accepts holes from NPB and electrons from AlQ, just like the AlQ layer in a conventional NPB/AlQ (emissive layer)/AlQ (electron-transport layer) structure.

In summary, an efficient 1.54 μm Si-based OLED with a structure of NTPS/NPB/ErQ/AlQ/Al is demonstrated. Its current and power efficiencies have been improved by two orders of magnitude, compared to the structure of *c*-Si/NPB/ErQ/Al, which is similar to that in a pioneering work.[6] The large improvement is attributed to (1) the NTPS anode controlling the hole injection, (2) the AlQ layer blocking the holes, and (3) the enhanced infrared light out coupling.

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