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1.54 μ m electroluminescence from erbium (III) tris(8-hydroxyquinoline) (ErQ)-based organic light-emitting diodes

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Organic light-emitting diodes have been fabricated using erbium tris(8-hydroxyquinoline) as the emitting layer and N, N'-diphenyl-N, N'-bis(3-methylphenyl)-1, 1'-biphenyl-4, 4'-diamine as the hole-transporting layer. Room-temperature electroluminescence was observed at 1.54 μ m due to intra-atomic transitions between the ${}^4I_{13/2}$ and ${}^4I_{15/2}$ levels in the Er³⁺ ion. These results suggest a possible route to producing a silicon-compatible 1.54 μ m source technology. © 1999 American Institute of Physics. [S0003-6951(99)02336-0]

Erbium-doped materials have for many years been the subject of much interest due to their application in optical fiber communications. The ${\rm Er}^{3+}$ ion has a sharp luminescence centered at 1.54 $\mu{\rm m}$ due to an intra-4f shell transition between the first excited state ($^4I_{13/2}$) and the ground state ($^4I_{15/2}$). Given that silicon is transparent at 1.54 $\mu{\rm m}$, it has been doped with erbium, often with other dopants such as oxygen and fluorine, with the hope of producing a siliconbased 1.54 $\mu{\rm m}$ emitter technology. There are, however, problems with this approach as the erbium-related luminescence tends to quench at room temperature.

Organic light-emitting diodes (OLEDs) have been the subject of much research since Tang and VanSlyke demonstrated electroluminescence from aluminum tris(8-hydroxyquinoline) (AlQ). Since then, considerable work has been done on improving OLED characteristics such as lifetime, brightness, and efficiency. In this letter, we report the fabrication of an OLED emitting electroluminescence at 1.54 μ m using erbium tris(8-hydroxyquinoline) (ErQ) as the emitting layer and N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) as the hole-transporting layer.

The ErQ was produced by mixing erbium (III) chloride in aqueous solution with 8-hydroxyquinoline in methanol. Following purification, it was placed into a boron nitride crucible in an UHV chamber for sublimation. First, 500 Å of TPD was evaporated onto cleaned indium-tin-oxide-coated glass (ITO), with a sheet resistance of 20 Ω/\Box , followed by 600 Å of ErQ. 1100 Å of aluminum was then evaporated to form the top electrode. The I-V characteristics were obtained using a Keithley 236 current-voltage source. The electroluminescence was dispersed in a 1 m spectrometer and recorded using a 0.5 µm Blaze grating with an S-20 photomultiplier for the "band-edge" luminescence and a 1 μ m Blaze grating with a liquid-nitrogen-cooled Ge detector for the erbium-related luminescence. The photoluminescence obtained has been described in a previous paper.³ All spectra were obtained at room temperature.

Figure 1 shows the "band-edge" electroluminescence

emitted under a driving voltage of 25 V along with the photoluminescence previously reported. While there is little change in the main peak positions of the two spectra, it can be seen that the electroluminescence does not have the higher-energy peak which appears in the photoluminescence spectra. The electroluminescence spectra peaks at ~600 nm and has a full width at half maximum (FWHM) of 127 nm. It was also considerably weaker in intensity than the electroluminescence, observed under the same operating conditions, from OLEDs with other group III chelates (such as AlQ) used as the emitting layer. Figure 2 shows the erbium-related electroluminescence, obtained under the same conditions as the "band-edge" electroluminescence, along with the photoluminescence for the same region. The two spectra are markedly different with the electroluminescence having a FWHM of \sim 33 nm compared to \sim 76 nm for the photoluminescence.

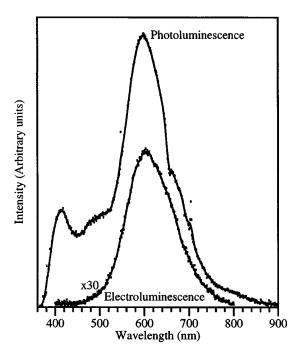


FIG. 1. The 300 K ''band-edge'' photoluminescence of ErQ excited using the 351 nm line from an argon-ion laser and the ''band-edge'' electroluminescence obtained from an ITO/TPD(500 Å)/ErQ(600 Å)/Al OLED. Note that the solid lines are a guide to the eye.

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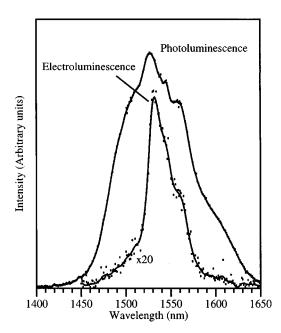


FIG. 2. The 300 K erbium-related photoluminescence excited using the 457 nm line from an argon-ion laser and the erbium-related electroluminescence obtained from an ITO/TPD(500 Å)/ErQ(600 Å)/Al OLED. Note that the solid lines are a guide to the eye.

Furthermore, the main luminescence peak of the electroluminescence is at 1533 nm, which is close to the emission seen from erbium in other materials, this is significantly shifted from the peak of the photoluminescence of ErQ which is at 1525 nm. Close investigation of Fig. 2 does show the presence of a shoulder at \sim 1533 nm in the erbium-related photoluminescence and the peak at \sim 1558 nm is present in both spectra. The most obvious difference between these two spectra is that the photoluminescence has appreciable emission between 1450 and 1520 nm, which is very much reduced in the electroluminescence.

These differences between the photoluminescence and the electroluminescence may be indicative of different forms of ErQ. It is known that the Stark splitting of the energy levels in Er^{3+} depends on the local electric field seen by the atom. Furthermore, it is known that for AlQ there are two isomers of the molecule, the fac (C_{3v} symmetry) and mer (C_{2v} symmetry). It is possible that the material used for the photoluminescence, which has not been sublimed, could be a mixture of these two isomers resulting in emission from erbium atoms in different local environments. However, the sublimation required to form the OLED may have resulted in this material having a higher concentration of one of the isomers.

The differences in the high-energy side of the "bandedge" luminescence between the photoluminescence and electroluminescence may be due to the presence of these structural differences or to other impurities, and further work will be necessary to understand this.

Figure 3 shows the current–voltage characteristics for the device and the erbium-related electroluminescence intensity as a function of driving current. From the I-V characteristics it can be seen that the device turn-on voltage is 12 V. This high turn-on voltage is indicative of poor injection efficiencies and is probably caused by the use of aluminum

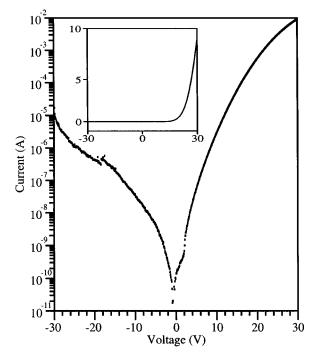


FIG. 3. Current-voltage characteristics for an ITO/TPD(500 Å)/ErQ(600 Å)/Al OLED. The inset shows the same data plotted on a linear scale with the vertical axis in mA.

as the contact electrode. There is no reason to suppose that this turn-on voltage cannot be reduced as has been achieved with AlQ-based diodes. The device shows excellent diode characteristics, however, and does not break down under reverse biases as high as -30 V (Fig. 3). We have performed some preliminary measurements of electroluminescence intensity against drive current and these indicate that for drive voltages up to 30 V we have not saturated the erbium-related emission, and so, it should be possible to obtain brighter luminescence. It should be noted, however, that our drive current at 30 V is an order of magnitude greater than that observed in TPD/AlQ diodes of similar geometry. These greater currents may be indicative that the ErQ is a better hole transporter than AlO, with the possibility that a number of the injected carriers are traversing the diode without recombination and are thus not contributing to the electroluminescence. Thus, by increasing the thickness of the ErQ layer or introducing hole-blocking layers, it may be possible to increase the emission intensity and maybe to saturate the erbium luminescence.

It has been demonstrated by Zhou *et al.*⁵ and Kim *et al.*⁶ that OLEDs can be fabricated using *p*-type doped silicon as the anode with TPD and AlQ as the organic layers. Given the results presented here, we suggest that this provides a possible route to producing a 1.54 μ m silicon-integrated emitter technology.

In conclusion, we have demonstrated 1.54 μ m room-temperature electroluminescence from OLEDs using ErQ as the emitting layer and have highlighted the possibility of a route to producing a silicon-based organic light-emitting diode operating at this wavelength.

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