Extended Abstracts of the 2008 International Conference on Solid State Devices and Materials, Tsukuba, 2008, pp. 628-629

P-10-15

Anomalous Temperature-dependent Behaviors of Electroluminescence Phenomena in the Disordered ITO/PEDOT/PFO/Ca/Al Polymer Light-emitting Diodes

Jen-Wei Teng, Jen-Cheng Wang, Yen-Hsun Lu, Tzer-En Nee*, and Gwo-Mei Wu

Institute of Electro-Optical Engineering and Department of Electronic Engineering, Chang Gung University
259 Wen-Hwa 1st Road, Kwei-Shan, Tao-Yuan, Taiwan, Republic of China.
Tel: +886-3-2118800 ext. 5791, Fax: +886-3-2118507, E-mail: neete@mail.cgu.edu.tw

1. Introduction

The past decades have been marked by significant advances in the construction of new light sources and screens. Recently, organic semiconductor devices have received much attention due to intensive applications of organic light-emitting diodes (OLEDs) in lightening, flat panel displays, and optically pumped organic thin films in stimulated emission [1,2]. However, in a statistical order–disorder view, the Berthelot-type microscopic model has successfully described the linear and the nonlinear temperature-dependent behaviors of many disordered systems, such as porous Sr_2CrReO_6 ferrimagnets, porous Si semiconductors, and CdS nanocrystalline thin films [3]. In this work, we reported the organic semiconductor devices of disorder materials concerning the thermal-related attributes arising from ITO/PEDOT/PFO/Ca/Al polymer light-emitting diode (PLEDs) structure. The characteristics of device have been studies as temperature- and excitation-dependent measurement of electroluminescence (EL). The temperature- and current-dependent EL spectra from the sample were measured over a temperature range from 300 to 20 K with current 0.5 mA and 0.1 mA and explained well by the localized carrier hopping and thermalization process. Furthermore, the thermal effect on the EL spectra with increasing driving current is also discussed in this work.

2. Experiment

Organic layers were deposited by spin coater onto a glass substrate coated with a patterned indium-tin-oxide (ITO) electrode. A hole injection layer PEDOT:PSS was spin coated onto ITO glass substrate. Next, the active luminescent polymer film PFO was spin coated onto PEDOT:PSS layer. Thereafter, the samples were transferred into thermal evaporation chamber that the electron injection layer, Ca (30 nm) and the Al cathode (~100 nm) were evaporated sequentially. For the temperature-dependent EL measurements, samples were mounted in a closed-cycle He cryostat to vary the sample temperature over a wide range (T = 20-300 K). Current-voltage (I–V) characteristics of the diodes were measured using a semiconductor parameter analyzer.

3. Results and discussion

Figure 1 shows the I-V characteristics of the device as a function of the ambient temperature. It is found that the values of V_F are 4.1 V, and 5.1 V for the sample with 0.1 mA at the temperatures of 20 K, and 300 K, respectively. Experimentally, both the V_F values and the dynamic resistances decreased with increasing temperature, indicating a change in the contact resistance between the metal and organic layers and the thermal effect.

In order to inspect the dependence of the dynamical carrier transport in the PLED, it is of interest to examine the radiative recombination of the confined electrons and holes at low temperature. The excitation-dependent output spectra for ITO/PEDOT/PF/Ca/Al PLED which operated at 0.1 mA and 0.5 mA over the temperature range of 20-300 K, are plotted in Figs. 2 (a) and (b). At room temperature, the MQW peak is observed around wavelength of 535 nm. All the intensities were normalized to the values observed at 300 K. When temperature is slightly decreased from 300 K, the luminescent intensities for the PLEDs with 0.1 mA and 0.5 mA efficiently increase and reach the maximum at 260 and 200 K, respectively. With further decrease of temperature down to 20 K, it is found that the device with 0.1 mA exhibits a smaller reduction of intensity, the device with 0.5 mA exhibits larger one. The observation of the reduction of the EL intensity at a low temperature is similar to experimental reports, where the abnormal behaviors manifested the peculiar radiative mechanisms of the nitride-based QW heterostructures by electrical excitation. There was a slow intensity-collapse rate, resulting from the suppression of the leakage carriers and the reduction of the thermal effect in the PLED structure, leading to the exhibition of a high spectral efficiency. The temperature dependence of the abnormal optical properties and the turning-point temperature, which is designated as T_t, are uniquely characterized [4]

$$T_t = \frac{M\Omega^2}{2\alpha^2 k} \qquad (1)$$

where k, M, and Ω are the Boltzmann constant, the inertia of the vibrating system, and the hopping frequency, respectively. α^{-1} is the extent of the carrier wave function. With carriers drifting coherently across the crystallite microbarriers, it was essential to observe a high T_t due to the predominance of the Berthelot-type process. As strong electron-electron scattering arising from a high excitation and a microcrystalline randomization, a high hopping frequency led to that the Berthelot-type process correspondingly dominated with a small T_t. Therefore, it has been that T_t decreases gradually with increasing driving current. When temperature is further decreased from turning point temperature to 80 K, the reduction of EL intensity is much faster and steeper for both current. That is,

electrically injected carriers are not efficiently captured and recombined in PLEDs diode when low temperature range. When temperature decreases below 80 K, EL intensity is until to saturation with material self characteristic.

Fig. 4 shows an Arrhenius plot of the integrated EL intensity of emission state for the PLED sample structure with driving current 0.1 and 0.5 mA, over a broad temperature range. The solid line in Fig. 4 represents the least-square fit of the above equation to the experimental data. It is obvious that the fitting is in fairly good agreement with the experimental result. The following expression is generally used to calculate the activation energy (E_A) in thermally activated processes [5]:

$$\mathrm{I(T)} = \mathrm{Rm} = \frac{\mathrm{P}}{1 + 2\dfrac{1 + U\beta_A/R}{U/R' + U_1\beta_B/R'}} \qquad (2)$$

where

$$\beta_i = \exp\left[-\frac{\mathrm{E}_i}{k\mathrm{T}}\right] \quad (i = \mathrm{A}, \mathrm{B})$$

In the Eq. (2), P is the excitation or pumping rate, R is the radiative recombination rate constant from the dots, R' is the nonradiative rate constant from the barrier, U and U_1 are the electron-heavy hole pairs and electron-light hole pairs trapping rate constant of organic material PF, respectively. I(T) is the temperature-dependent integrated EL intensity, k_B is the Boltzmann's constant. We obtain the activation energy values for the driving current 0.1 and 0.5 mA from the low-excitation Arrhenius plot in Fig. 3 are 126.6 and 112.2 meV. The smaller activation energy is expected to facilitate the redistribution of the thermal carrier overcoming the energy barrier and the rapid degradation of luminescence. This resulted in not only merely increasing the carrier confinement, but also enhancing the quantum efficiency of PLED structures with lower excitation.

4. Conclusions

In this study, the organic semiconductor devices of disorder materials have been systematically investigated. It is found that the higher turning-point temperature shows a correlation with carrier hopping frequency and the degree of EL intensity with activation energy. Not only the abnormal emission energy evolutions are found to be temperature dependent, but also the acceptable values of the Berthelot-type temperature lie within 200 and 260 K. Therefore, it is observed the anomalous temperature behavior of EL spectra from the sample are investigated by the localized carrier hopping and thermalization process and exhibited to be in a fair agreement with the experimental results.

References

[1] C. W. Tang et al., Appl. Phys. Lett. **51**, 913 (1987).
[2] L. S. Hung et al., Appl. Phys. Lett. **78**, 544 (2001).
[3] T. Z. Nee et al., J. Crystal Growth. **287**, 468(2006).
[4] George C. John et al., Phys. Rev. B **54**, 7 4416 (1996).
[5] W. H. Jiang et al., J. Appl. Phys., **88**, 2529 (2000).

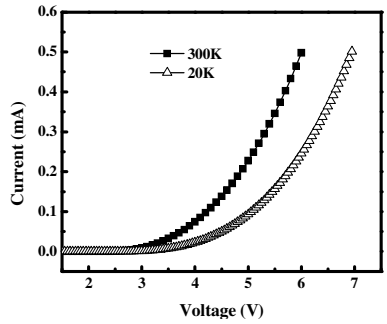

Fig. 1 Current-voltage curves of PLED device at 20 and 300 K.

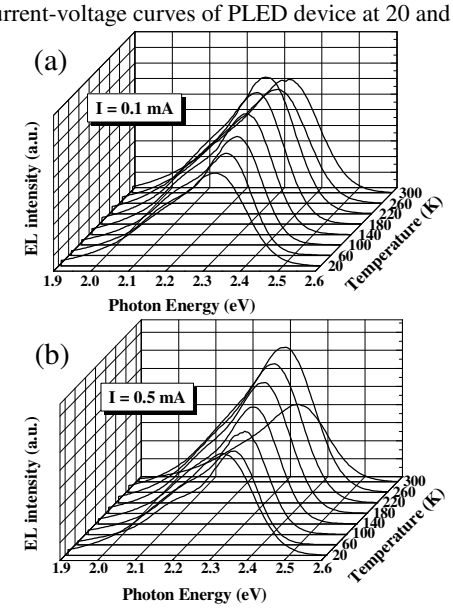

Fig. 2 Temperature-dependent electroluminescence spectra of samples with driving current (a) 0.1 mA (b) 0.5 mA.

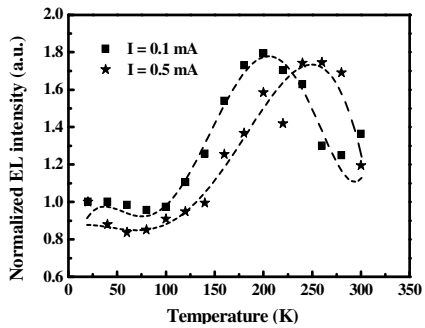

Fig. 3 The temperature-dependent of normalized EL intensity for PLED device with injection current of 0.1 mA and 0.5 mA.

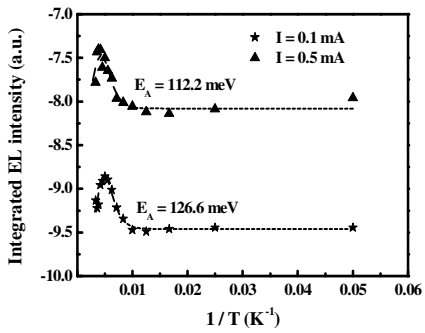

Fig. 4 Temperature dependence of the EL integrated intensity from the sample with driving current 0.1 mA and 0.5 mA.