Confirmation on origin of primary electron in solid state cathodoluminescence*

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(Received 2 November 2009; revised manuscript received 25 November 2009)

This paper utilizes the brightness-voltage waveform curve to investigate the primary electron in solid state cathodoluminescence. The results indicate that the primary electron is from the interface state of SiO_2/MEH-PPV (Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]) under the lower electric field which contributes to the 580-nm emission. With increasing the electric field, the 405-nm emission is obtained, and under this condition, the origin of the primary electron is mainly from tunneling.

Keywords: primary electron, solid state cathodoluminescence, organic–inorganic heterostructure

PACC: 7860F, 7220D, 7280L

1. Introduction

Solid-state cathodoluminescence (SSCL) is a kind of luminescence mechanism in the organic–inorganic heterostructure light emitting diodes family. Compared with traditional cathodoluminescence ray tube (CRT), the process of electron acceleration is in the SiO_2 layer rather than in vacuum.[1−4] The luminescence mechanism of SSCL is confirmed,[1] the origin of the long wavelength is believed to be exciton luminescence and the exciton is from the impact excitation. The short wavelength emission comes from the direct band–band radiative recombination of carriers caused from exciton dissociation.[5,6]

In the luminescence of SSCL, the impact excitation is an important process which is dominated by the primary electron,[7−10] but there is no direct evidence to identify the origin of the primary electron. So the confirmation on the origin of the primary electron is essential to optimize and improve the SSCL.

In this paper, the origin of the primary electron in SSCL is confirmed by means of brightness-voltage waveform curves.

2. Experimental

The devices ITO/MEH-PPV (120 nm)/Al (device A), and ITO/SiO_2 (200 nm)/MEH-PPV (120 nm)/SiO_2 (200 nm)/Al (device B) have been fabricated. The SiO_2 layer was prepared by electron beam evaporation (EVA450, Alliance Concept Co. Ltd., France). In this process, the substrate temperature was kept at 160 °C, the vacuum was 2×10^{-6} Torr (1 Torr=133 Pa) and the growth rate was 0.3 nm/s and the thickness was controlled by a quartz crystal thickness monitor. The MEH–PPV was dissolved in chloroform and then it was fabricated by the spin-coating method. The top Al electrode was prepared by thermal evaporation under the vacuum 10^{-6} Torr. The electroluminescence was measured by SPEX Fluorolog-3 spectrometer. The brightness-voltage waveform curves were obtained by an oscillograph (Tektronix TDS 540D).

3. Results and discussion

Figure 1(a) shows the normalized electroluminescence spectrum of device A driven by AC voltage of 500 Hz, and there is only one peak located at 580 nm. The electroluminescence spectrum of device B driven

*Project supported by the Doctoral Foundation of University of Jinan (Grant No. XBS0845), the Key Laboratory of Luminescence and Optical Information (Beijing Jiaotong University), Ministry of Education, China (Grant No. 2010LOI01), the Natural Science Foundation of Shandong Province, China (Grant Nos. Y2008A21 and SZR0704), the Scientific Research Foundation of the Education Department of Shandong Province, China (Grant No. J08LI12).

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by AC voltage of 500 Hz is shown in Fig. 1(b), and two emission peaks, the long wavelength 580 nm and short wavelength 405 nm, are obtained. The spectrum is characterized by the appearance of short wavelength emission after the applied voltage is over a certain value. Figure 1(c) shows that there is only the long wavelength emission when the voltage is low. With increasing applied voltage, both long and short wavelength emissions can be obtained, and then the long wavelength peak disappears completely and the short wavelength is achieved only.

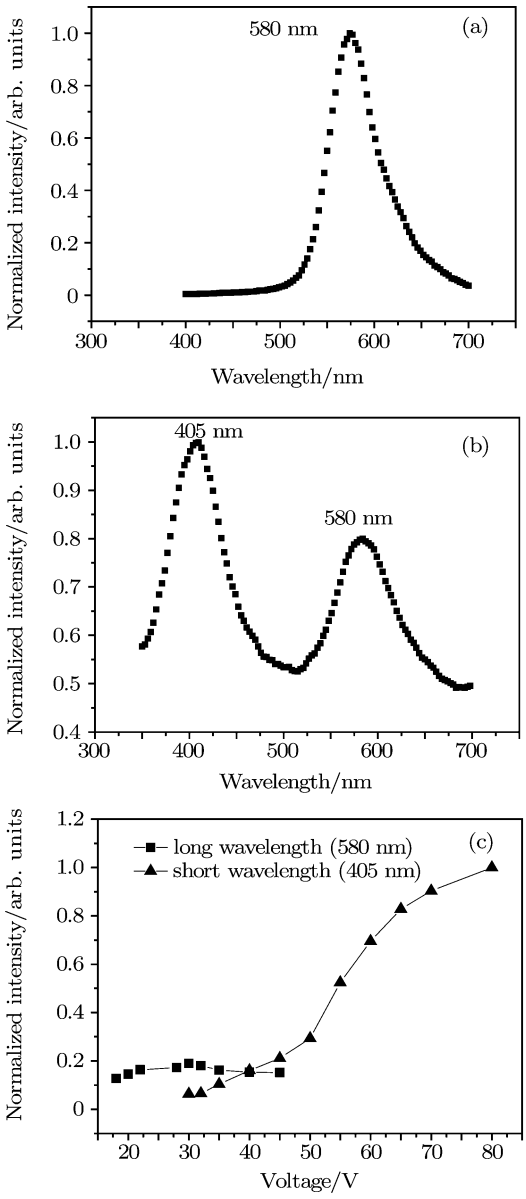


Fig. 1. Electroluminescence spectrum of the devices A (a), B (b), and (c) electroluminescence intensity of wavelength versus voltage of the device B.

To our knowledge, the origin of the long wavelength is exciton luminescence caused from hot electron impact excitation. The short wavelength emission comes from the direct band–band radiative recombination of carriers caused from exciton dissociation. In this luminescence mechanism, the process of impact excitation is dominated by the primary electron.

In order to assure the origin of the primary electron, we apply the luminescence-voltage waveform curve. Figure 2(a) is the luminescence–voltage waveform curve for the long wavelength emission 580 nm of device B under sinusoidal AC voltage 20 V. The emission peak is detectable in both directions and the intensities of both directions are similar (ITO electrode connected with the anode). For comparison, the luminescence–voltage waveform curve of device A for the 580-nm emission under AC voltage 13 V is obtained (Fig. 2(b)). There is only one emission peak when the ITO is anode. This implies that the emission mechanism of the 580-nm emission in these two devices is completely different. Compared with traditional injection luminescence in device A, we find that the long wavelength emission is exciton luminescence in device B caused by hot electron impact excitation. Because there is no obvious difference in long wavelength luminescence intensity between forward and inverse voltages in device B, that is to say the effect of potential barrier on the origin of primary electron can be neglected. So the primary electron comes from the interface state of SiO_2/MEH-PPV. In injection mechanism, however, potential barriers are the dominant factor in the luminescence of device A. So the luminescence intensity is different when the device is driven by forward and inverse voltage.

Figure 2(c) shows the luminescence–voltage waveform curve for short wavelength emission 405 nm of device B under AC voltage 50 V. The emission is weak under inverse voltage in comparison with forward voltage. Because the impact excitation is the important process in the SSCL luminescence mechanism in device B, so the luminescence intensity depends on the number of the primary electron. Figure 2(c) indicates that the number of the primary electron is different for forward and inverse voltage. The energy band diagram of all material used is shown in Fig. 2(d). It can be seen that there is a 1.0 eV for electron injection from ITO to SiO_2, but the injection barrier is 0.6 eV from Al to SiO_2, so the electron injection under forward voltage is easier than that under inverse voltage, which is in agreement with the luminescence intensity. So with the increase of the electric field, the origin of primary electron is mainly from tunneling.

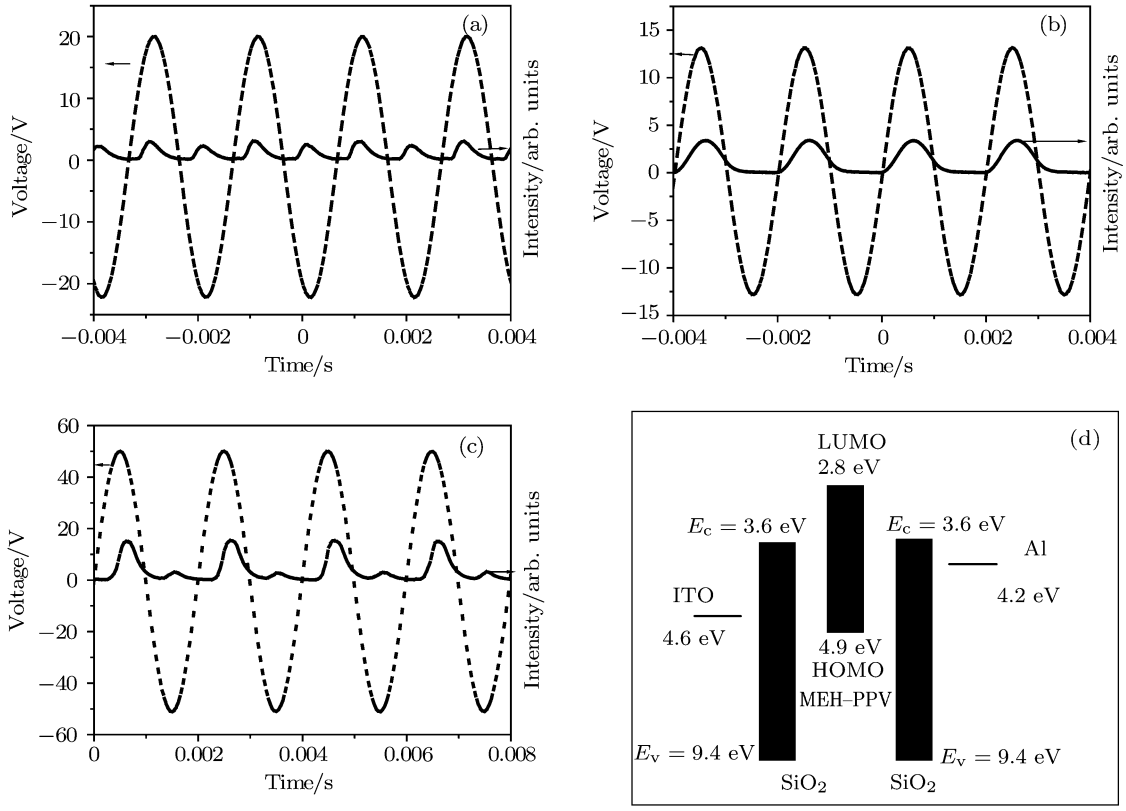


Fig. 2. (a) Luminescence–voltage waveform curve of device B for the 580-nm emission under AC 20 V. (b) Luminescence–voltage waveform curve of device A for the 580-nm emission under AC 13 V. (c) Luminescence–voltage waveform curve of device B for the 405-nm emission under AC 50 V, (d) The energy band diagram of SiO_2, MEH–PPV, ITO and Al.

4. Conclusions

In summary, based on traditional SSCL structure, the origin of the primary electron is studied by means of a brightness–voltage waveform curve. The results indicate that the primary electron is from the interface state under lower electric field, and tunneling is the main source of primary electron under increasing electric field.

References

[1] Xu X R 2007 *J. Lumi.* **122–123** 593
[2] Qu C, Xu Z, Zhang F J, Ma X G and Xu X R 2009 *J. Appl. Phys.* **106** 023111
[3] Qu C, Xu Z, Teng F and Xu X R 2003 *Chin. Phys. Lett.* **20** 1144
[4] Zhang F J, Zhao S L, Xu Z, Huang J Z and Xu X R 2007 *Chin. Phys.* **16** 1464
[5] Li Y, Zhao S L, Xu Z, Zhang F J, Huang J Z, Song L and Ou Y P 2007 *Acta Phys. Sin.* **56** 5526 (in Chinese)
[6] Huang J Z, Xu Z, Zhao S L, Zhang Z, Wang P J and Zheng Z S 2009 *Physica* B: *Condensed Matter* **404** 1595
[7] Zhang F J, Xu Z, Teng F Yang S Y, Lou Z D, Liu L, Meng L J, Hou Y B, Wang Y S and Xu X R 2006 *J. Lumi.* **117** 90
[8] Zhang F J, Xu Z, Teng F, Liu L and Meng L J 2005 *Mater. Sci. Eng.* B **123** 84
[9] Huang J Z, Xu Z, Zhao S L, Li Y, Yuan G C, Wang Y S and Xu X R 2007 *Physica* B: *Condensed Matter* **394** 115
[10] Qu C, Xu Z, Teng F, Qian L, Yu W G, Quan S Y and Xu X R 2004 *Acta Phys. Sin.* **53** 4387 (in Chinese)