Extended Abstracts of the 2006 International Conference on Solid State Devices and Materials, Yokohama, 2006, pp. 74-75

D-1-1 (Invited)

Highly efficient carrier injection and transport in Organic Light Emitting Diodes

Toshinori Matsushima and Chihaya Adachi

Core Research for Evolutional Science and Technology Program,
Japan Science and Technology Agency, 1-32-12 Higashi, Shibuya, Tokyo 150-0011, Japan
Center for Future Chemistry, Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan

1. Introduction

Recently, organic light-emitting diodes (OLEDs) have been attracting considerable attention due to their high potential for future display systems, i.e., thin, flexible, and low-cost flat panel displays. For this purpose, high energy conversion efficiencies have been required and various optimizations of materials and device structure to decrease driving voltages have been developed. Further, organic semiconductor laser diodes (OSLDs), which can be classified as post-OLED applications, have emerged as a challenging research subject in organic optoelectronic devices.[1-3] To realize OSLD, a current density greater than several kA/cm^2 have to be injected into an organic thin film with application of practical voltage.

In this study, we demonstrate high performance organic semiconductor devices having very low voltage driving using efficient electron and hole injection layers. Further, using small active device area, we demonstrate extremely high density carrier injection and transport into organic thin films.

2. Small area device

In a previous study, we fabricated a multi-layered OSLD structure sandwiched with transparent indium tin oxide (ITO) anode and cathode, which allows not only optical but also electrical pumping.[3] With this device structure, we observed amplified spontaneous emission (ASE) with a threshold energy of E_{th} = 12.8 μJ/cm^2 under optical pumping. From the optical threshold energy, we estimated that the threshold current density required for the ASE under the electrical pumping was approximately J = 3.8 kA/cm^2. In order to inject and transport such high current density, we previously demonstrated that a decrease in active device areas and the use of high thermal conductivity substrates enable us to realize such high current densities.[4-6] For example, a CuPc thin-film device with a small active area of S = 1962 μm^2 (a circular cathode with a radius of r = 25 μm) and a high thermal conductivity silicon substrate had a steady-state high current density of J_{MAX} = 12 kA/cm^2.[5] In these devices, the active device areas were defined by the overlap of an anode and a metal cathode patterned by a shadow mask. Conventional shadow masking, however, makes it difficult to practically reduce the active areas to less than S = 1962 μm^2 (r = 25 μm) due to the incomplete deposition of evaporated materials through the small openings in the shadow mask. Therefore, we introduced a photolithography and electron-beam techniques that can more straightforwardly reduce and precisely control the active areas than the shadow-masking technique.[6] An insulating photo-resist film was spin-coated on an ITO substrate, and "tiny holes" connecting it to the ITO surface were patterned in the photo-resist film. The active areas were defined well by the hole sizes in the photo-resist film, and small active areas from S = 625 to 0.04 μm^2 were controlled.

We would like to stress the importance of understanding carrier injection and transport mechanisms in organic thin films in a wide current density range from nA/cm^2 to kA/cm^2 and the technical importance of fabricating small devices. Although much research has been done on the carrier injection and transport mechanisms in organic thin films (for example, Refs. 7, 8, and 9), the mechanisms are not fully understood. In particular, the maximum current density has been limited to as low as $J \approx$ 10 A/cm^2 because most organic thin-film devices encounter irreversible breakdowns at current densities lower than J = 10 A/cm^2, resulting in difficulties in analyzing the detailed carrier injection and transport mechanisms under current densities higher than J = 10 A/cm^2. In this report, we demonstrate that CuPc thin-film devices having the smallest areas of S = 0.04 μm^2 sustain high current density operation up to J_{MAX} = 6.35 MA/cm^2. Further, we observed a trap-free space-charge-limited current at a current density higher than $J \approx$ 80 kA/cm^2.

3. Unique J-V characteristics

The J-V curves of the CuPc devices with the different active areas, S = 625, 117, 34.6, and 7.9 μm^2, under dc bias operation are shown in Fig. 1. The J_{MAX} and V_{MAX} values are strongly dependent upon the active device areas, whereas all of the

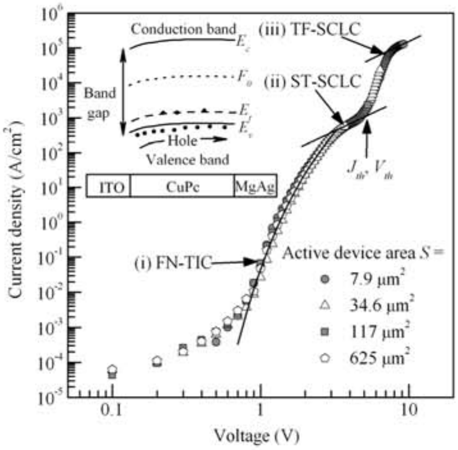

Fig. 1: Current density versus voltage curves for CuPc thin-film devices with different active device areas, S = 625, 117, 34.6, and 7.9 μm². Experimental curves are fitted with theoretical single-carrier injection equations (solid lines) of (i) FN-TIC, (ii) ST-SCLC, and (iii) TF-SCLC models.

CuPc devices follow the same J-V curves. Although the largest device, with S = 625 μm², had J_{MAX} = 574 A/cm² at V_{MAX} = 3.7 V, the J_{MAX} and V_{MAX} values markedly increased as the active device areas were decreased from S = 625 to 7.9 μm², and the J_{MAX} value in the smallest device, with S = 7.9 μm², reached J_{MAX} = 128 kA/cm² at V_{MAX} = 9.2 V, which is ≈ 200 times higher than that in the largest device, with S = 625 μm². Using electron-beam lithography technique, further, we fabricated a copper phthalocyanine (CuPc) thin-film device having a very small active device area on a high thermal conductivity silicon substrate, and demonstrate an extremely high breakdown current density of J_{MAX} = 6.35 MA/cm² with S = 0.04 μm² (Fig. 2). This high current density injection and transport reveal that current density-voltage characteristics of the CuPc device can be categorized into the four charge-carrier transport regions, i.e., ohm current, shallow-trap space-charge-limited current (SCLC), trap-free SCLC, and two-carrier injection current processes, under current densities from nA cm^{-2} to MA cm^{-2}.

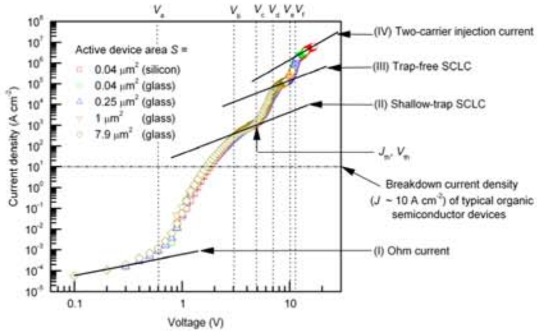

Fig. 2: Steady-state current density-voltage (J-V) characteristics of CuPc thin-film devices with different active device areas (S = 7.9, 1, 0.25, and 0.04 μm²) and substrates (glass and silicon).

References

1. V. G. Kozlov, V. Bulovic, P. E. Burrows, and S. R. Forrest, Nature **389**, 362, (1997).
2. T. Aimono, Y. Kawamura, K. Goushi, H. Yamamoto, H. Sasabe, and C. Adachi, Appl. Phys. Lett. **86**, 071110, (2005).
3. H. Yamamoto, T. Oyamada, H. Sasabe, and C. Adachi, Appl. Phys. Lett. **84**, 1401, (2004).
4. W. Yokoyama, H. Sasabe, and C. Adachi, Jpn. J. Appl. Phys., Part 2, **42**, 1353, (2003).
5. H. Yamamoto, H. Kasajima, W. Yokoyama, H. Sasabe, and C. Adachi, Appl. Phys. Lett. **86**, 083502, (2005).
6. C. I. Wilkinson, D. G. Lidzey, L. C. Palilis, R. B. Fletcher, S. J. Martin, X. H. Wang, and D. D. C. Bradley, Appl. Phys. Lett. **79**, 171, (2001).
7. P. E. Burrows, Z. Shen, V. Bulovic, D. M. McCarty, S. R. Forrest, J. A. Cronin, and M. E. Thompson, J. Appl. Phys. **79**, 7991, (1996).
8. R. D. Gould, Thin Solid Films **125**, 63, (1985).
9. I. H. Campbell, D. L. Smith, C. J. Neef, and J. P. Ferraris, Appl. Phys. Lett. **75**, 841, (1999).
10. N. Tessler, N. T. Harrison, D. S. Thomas, and R. H. Friend, Appl. Phys. Lett. **73**, 732, (1998).
11. J. C. Scotto, J. H. Kaufman, P. J. Brock, R. DiPietro, J. Salem, and J. A. Goitia, J. Appl. Phys. **79**, 2745, (1996).
12. Z. Bao, A. J. Lovinger, and A. Dodabalapur, Appl. Phys. Lett. **69**, 3066, (1996).
13. T. Okuda, S. Shintoh, and N. Terada, Appl. Phys. Lett. **96**, 3586, (2004).
14. M. A. Lampert and P. Mark, *Current Injection In Solids* (ACADEMIC, New York, 1970).