Alignment-free process for asymmetric contact electrodes and their application in light-emitting organic field-effect transistors

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We developed an alignment-free process for asymmetric contacts of Au and Al and applied it to light-emitting organic field-effect transistors. Because electrons were injected efficiently from Al contacts, the emission intensity and onset voltages for light were significantly better than those in a device with conventional Au/Cr contacts. Moreover, a device with 1 μm channel length asymmetric contacts of Au and Al showed about 50 times higher current than that of the device with conventional Au/Cr contacts. This significant improvement can be ascribed to both dual space-charge formation of holes and electrons and low carrier injection barriers. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2839895]

Simultaneously injecting holes and electrons into the semiconducting layers of organic field-effect transistors (OFETs) enables us to observe light emissions.[1–13] Such light-emitting OFETs (LE-OFETs) are attractive for studying fundamental optoelectronic characteristics in organic semiconducting films and for practical applications, especially for active matrix displays,[8] because LE-OFETs have two functions of light emissions and current switching in single devices.

Two different types of LE-OFETs have been reported. One is unipolar, where either holes or electrons accumulate in the channel and where the opposite carriers are injected by a strong electric field between source and drain contacts.[1–7] The other is ambipolar, which accumulates both charges of holes and electrons simultaneously.[9–13] Although the emission efficiency in ambipolar LE-OFETs is higher than that in unipolar ones, their electrical and emission behaviors are not suited for use in active matrix displays because their emission intensities do not depend simply on the drain currents. However, because emission intensity in the unipolar LE-OFETs is more directly correlated with the drain current, they provide a great advantage when we consider their application to displays.

Some key reports indicated that the unipolar LE-OFETs had improved emission efficiency and intensity. Sakanoue *et al.* found using the asymmetric contacts of Au and Al (Au–Al contacts) significantly improved hole and electron injections, leading to more efficient light emission.[4] Furthermore, Oyamada *et al.* found that the strong electric field between the source and drain contacts was crucial for enhancing electron injections.[6] We decided to combine these techniques. However, preparing LE-OFETs with short channel length asymmetric contacts is difficult because the preparation techniques reported so far require strict alignment of the photomask or metal shadow mask.[4,7]

For this study, we developed an alignment-free process to prepare asymmetric contacts by using a photoresist as a shadow mask for the angle deposition. LE-OFETs with these contacts can significantly improve the emission intensity and efficiency, and it can reduce onset voltage for light emissions compared to that with conventional Au/Cr contacts.

Figure 1(a) shows the preparation process for the Au–Al contacts in this work. Thermally oxidized SiO_2 (300 nm)/n^{++}Si was used for FET substrates. Conventional double-layer photolithography was carried out to determine the interdigital source and drain electrodes. The thickness of the photoresist was carefully controlled at 3.2 μm. Then, the specimens were introduced into a vacuum chamber to deposit Al and Au. First, Al was deposited with an angle of 60° normal to the substrates. Next, Au was successively deposited with an angle of −60°. The thicknesses were 30 nm for each metal. Because the patterned photoresist worked as a shadow mask, a single layer of Al was formed on one side of the edge of the electrodes, and on the other side, a single layer of Au was also formed. Figure 1(b) shows an optical micrograph of the electrode. We can see a clear asymmetric structure of Au and Al even when the channel length was as short as 1 μm. The conventional electrodes of Au (30 nm)/Cr(2 nm) were also prepared for comparison. The channel lengths of the device prepared in this study were 10 and 1 μm, and the channel widths in all devices were 80 mm. The FET substrates were treated with UV/O_3 and hexamethyldisilazane. 4,4′-bis[(*E*)-2-(4-hexylphenyl)vinyl]biphenyl (BSBP-C6), [Fig. 1(c)] was used as the active layers of the LE-OFETs because BSBP derivatives balanced both high mobility and high photoluminescence efficiency in a neat film.[11] It was sublimated three times and vacuum-deposited onto FET substrates under a vacuum of 1×10^{-6} Pa. Their thickness and rate were controlled at 30 nm and 2 nm/min, respectively. The completed devices were then transferred into the measurement chamber through the air. Electrical properties were obtained with a semiconductor device analyzer (Agilent B1500) under the vacuum of 1×10^{-3} Pa. Emission intensities were detected

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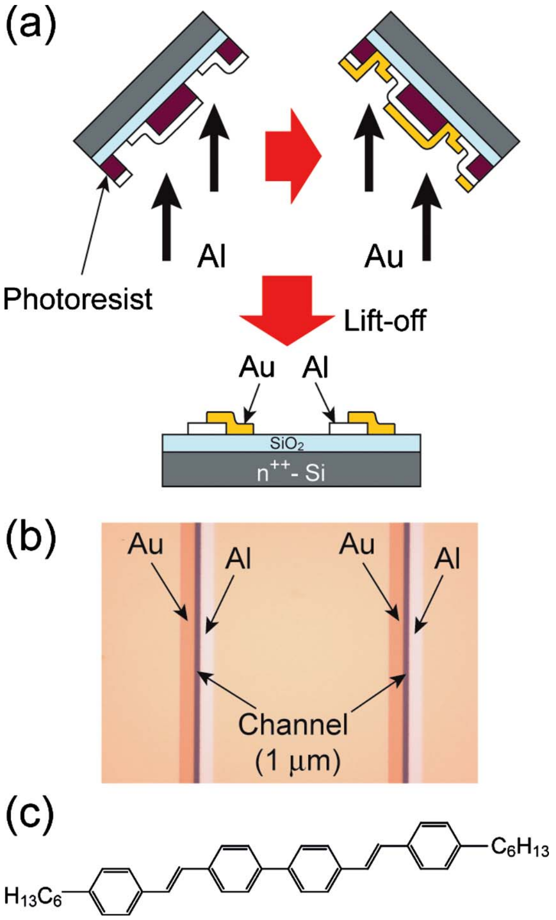

FIG. 1. (Color online) Preparation process for asymmetric contacts onto SiO$_2$/Si substrate (a). Optical micrograph of Au–Al contacts with channel length of 1 μm (b). Molecular structure of BSBP-C6 (c).

with a calibrated Si photodiode (Newport 818UV) through a quartz window of the chamber.

First, we mention the LE-OFET characteristics of the devices with the long channel length of 10 μm. Figures 2(a) and 2(c) show the FET and emission characteristics with the conventional Au/Cr contacts, respectively. The device showed p-type unipolar semiconducting behavior, and the field-effect hole mobility in this device was calculated to be 0.035 cm^2/V s. Weak emissions were observed at high drain and gate voltage ranges. Electron injections that lead to light emissions in p-type LE-OFETs with Au/Cr contacts occurred due to strong local electric fields in the drain contacts induced by pinch-off formation.[3,5] Therefore, the light emissions were mainly observed at saturation regions. On the other hand, the light emission was observed even from linear regions in the case of the device with Au–Al contacts, as Fig. 2(b) and 2(d) indicate. Thus, the electrons can be injected without a strong local electric field at the drain contacts because the work function of the Al electrode is low enough to inject electrons with low drain voltage. The onset drain voltage for the light emissions was about −10 V, and the shape of the emission characteristics was similar to the electrical ones. These behaviors gave us a relatively linear relationship between the drain current and emission intensity, as was demonstrated with 2–ethylhexyloxy-p-phenylenevinylene (MEH-PPV)-based LE-OFETs.[4] However, the electron injections in this device were still affected by the formation of the pinch-off. This behavior was seen as steep increases in the light intensity of the high drain voltage region in Fig. 2(d).

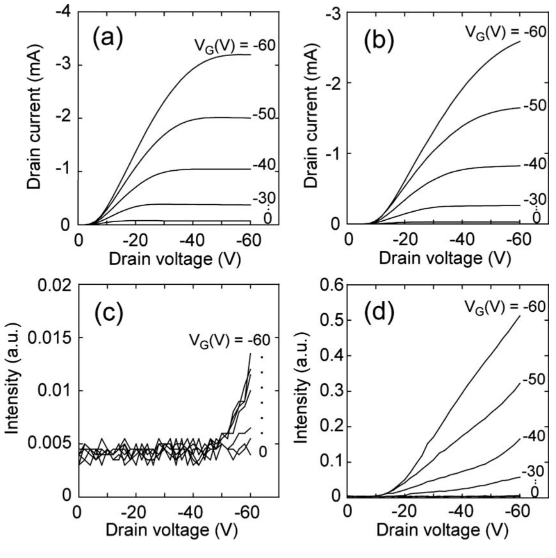

FIG. 2. FET [(a) and (b)] and emission characteristics [(c) and (d)] in 10 μm channel length LE-OFETs. (a) and (c) show the results for a device with Au/Cr contacts. (b) and (d) show the results for a device with Au–Al contacts.

The luminance intensity, 3 cd/m^2, was obtained in the device with Au–Al contacts at the drain and gate voltage of −60 V by a luminance meter. We note that the emission region was less than 1 μm, while the widths of electrodes were 50 μm. The apparent luminance would be improved by using thinner electrodes. Furthermore, the effective channel width of the device with Au–Al contacts was half compared to the device with Au/Cr contacts. The reason is that the Al and Au sides of our interdigital electrodes for the source and drain, respectively, were not suitable both for hole and electron injections.

Figures 3(a) and 3(b) show the FET characteristics of the 1 μm channel length LE-OFETs with Au/Cr and Au–Al contacts, respectively. The device with Au/Cr contacts showed p-type unipolar behavior with clear saturation behaviors. No light emissions were observed in this bias region due to a lower drain current than the 10 μm channel length device with AuCr contacts. However, in the device with 1 μm channel length Au–Al contacts, we observed no saturation behaviors in the FET characteristics and observed light emissions with a low onset drain voltage of −5 V, as shown in Fig. 3(c). The shape of the emission characteristics was almost the same as that of the electrical characteristics. This indicates that the external quantum efficiency was independent of applied drain and gate voltages and it was about 3×10^{-4}%. Moreover, we found that the drain current in the device with Au–Al contacts was about 50 times higher than that with Au/Cr contacts, even when the effective channel width was half.

Such higher current in emitting devices was also observed in the single layer organic light-emitting diode (OLED) structures.[14,15] The current in conventional OLEDs (double injection devices) usually does not correspond to the sum of the current in hole-only devices and electron-only devices. The current in double injection devices is sometimes more than two orders of magnitude higher than that in the hole-only or electron-only devices. Although this behavior is not yet understood clearly, it indicates that the formation of dual space charges of electrons and holes in the semicon-

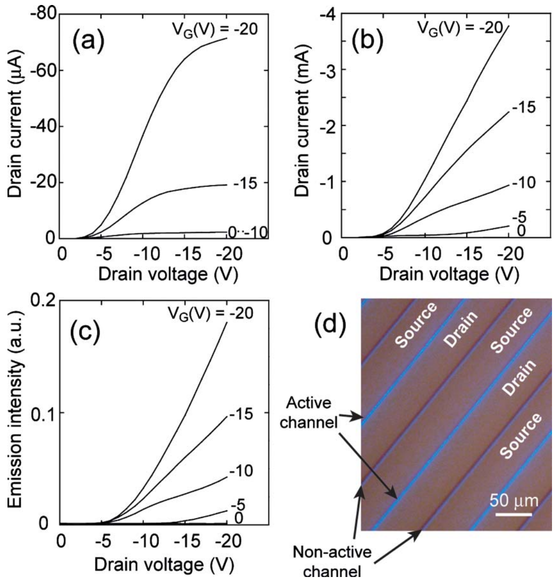

FIG. 3. (Color online) FET [(a) and (b)] and emission characteristics (c) in 1 μm channel length LE-OFETs. (a) shows the results for a device with Au/Cr contacts. (b) and (c) show the results for a device with Au–Al contacts. Optical micrograph of device with 1 μm channel length Au–Al contacts operating at drain and gate voltages of −30 V (d). Blue emission was observed in the channel where source contact was Au and drain was Al (active channel), whereas no light emission was observed in the channel where source was Al and drain was Au (nonactive channel).

ducting layers plays an important role in enhancing opposite carrier injections.[14] Our LE-OFETs with 1 μm channel length Au–Al contacts would contribute highly to dual space-charge-limited current because the drain current showed no saturation behaviors. Since the other LE-OFETs we prepared showed clear saturation behaviors, a combination of asymmetric Au–Al contacts and a short channel length of 1 μm causes higher drain current; low injection barriers allowed simultaneous injections of holes and electrons, and their space-charges enhanced opposite carrier injections in the short channel length device.

Figure 3(d) shows an optical micrograph of the device with Au–Al contacts driven at drain and gate voltages of −30 V. Blue emissions were observed at the alternative edges of the interdigital electrodes because half of the channels were not active as was previously mentioned. Thus, the work functions of the contacts are clearly very important even for short channel length LE-OFETs.

In summary, we developed an alignment-free process for preparing asymmetric Au–Al contacts. The LE-OFETs with this electrode significantly improved emission intensity and efficiency, and they reduced onset drain voltage for light emissions because the electron injection barrier was lowered by using Al for drain contacts. Furthermore, a device with 1 μm channel length Au–Al contacts showed about 50 times higher drain current compared to that with 1 μm channel length Au/Cr contacts. This behavior can be ascribed to the contributions of the dual space charges of holes and electrons and low injection barriers for both carriers. The asymmetric contacts with short channel length are also interesting for studying carrier injection and extraction processes at organic/metal interfaces.

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[1] A. Hepp, H. Heil, W. Weise, M. Ahles, R. Schmechel, and H. von Seggern, Phys. Rev. Lett. **91**, 157406 (2003).
[2] T. Sakanoue, E. Fujiwara, R. Yamada, and H. Tada, Appl. Phys. Lett. **84**, 3037 (2004).
[3] C. Santato, R. Capelli, M. A. Loi, M. Murgia, F. Cicoira, V. A. L. Roy, P. Stallinga, R. Zamboni, C. Rost, S. F. Karg, and M. Muccini, Synth. Met. **146**, 329 (2004).
[4] T. Sakanoue, E. Fujiwara, R. Yamada, and H. Tada, Chem. Lett. **34**, 494 (2005).
[5] T. Oyamada, H. Sasabe, C. Adachi, S. Okuyama, N. Shimoji, and K. Matsushige, Appl. Phys. Lett. **86**, 093505 (2005).
[6] T. Oyamada, H. Uchiuzou, S. Akiyama, Y. Oku, N. Shimoji, K. Matsushige, H. Sasabe, and C. Adachi, J. Appl. Phys. **98**, 074506 (2005).
[7] J. Reynaert, D. Cheyns, D. Janssen, R. Müller, V. I. Arkhipov, J. Genoe, G. Borghs, and P. Heremans, J. Appl. Phys. **97**, 114501 (2005).
[8] M. Muccini, Nat. Mater. **5**, 605 (2006).
[9] C. Rost, S. Karg, W. Riess, M. A. Loi, M. Murgia, and M. Muccini, Appl. Phys. Lett. **85**, 1613 (2004).
[10] J. Zaumseil, R. H. Friend, and H. Sirringhaus, Nat. Mater. **5**, 69 (2006).
[11] T. Sakanoue, M. Yahiro, C. Adachi, H. Uchiuzou, T. Takahashi, and A. Toshimitsu, Appl. Phys. Lett. **90**, 171118 (2007).
[12] E. C. P. Smits, S. Setayesh, T. D. Anthopoulos, M. Buechel, W. Nijssen, R. Coehoorn, P. W. M. Blom, B. de Boer, and D. M. de Leeuw, Adv. Mater. (Weinheim, Ger.) **19**, 734 (2007).
[13] T. Takahashi, T. Takenobu, J. Takeya, and Y. Iwasa, Adv. Funct. Mater. **17**, 1623 (2007).
[14] I. D. Parker, J. Appl. Phys. **75**, 1656 (1994).
[15] K. Murata, S. Cina, and N. C. Greenham, Appl. Phys. Lett. **79**, 1193 (2001).