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TECHNICAL NOTE

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Use of laser drilling in the manufacture of organic inverter circuits

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Abstract Inverter circuits have been made by connecting two high-quality pentacene field-effect transistors. A uniform and pinhole-free 900 nm thick polyimide gate-insulating layer was formed on a flexible polyimide film with gold gate electrodes and partially removed by using a CO_2 laser drilling machine to make via holes and contact holes. Subsequent evaporation of the gold layer results in good electrical connection with a gold gate layer underneath the gate-insulating layer. By optimization of the settings of the CO_2 laser drilling machine, contact resistance can be reduced to as low as 3 Ω for 180 μm square electrodes. No degradation of the transport properties of the organic transistors was observed after the laser-drilling process. This study demonstrates the feasibility of using the laser drilling process for implementation of organic transistors in integrated circuits on flexible polymer films.

Keywords Organic transistor · Laser drilling process · Inverter circuit · Pentacene

Organic field-effect transistors have attracted much attention because of their potential application in low-cost large-area flexible electronics, which would fit well with radio-frequency identification tags [1, 2], displays [3, 4], and particularly large-area sensors [5, 6]. Much progress has been made in the past few years and mobility exceeding 1 $cm^2 V^{-1} s^{-1}$ and an on/off ratio of 10^6 have been reported for thin-film transistors of evaporated pentacene [4, 7–11]. We have recently prepared highly mechanically flexible transistors with a mobility of 1 $cm^2 V^{-1} s^{-1}$ by using polyimide gate insulators on plastic films [12]. Much of this excellent performance has been demonstrated by using discrete organic transistors without device separation.

To implement organic transistors for integrated circuits, it is essential to make patterns of gate-insulating layers, in particular to make via holes through gate-insulating layers which make an electrical connection between gate electrodes and source/drain electrodes. Although some pioneering work with inkjet printing [13] and other methods has been reported [14], this technology requires precise control of viscosity and careful choice of solvents and is, therefore, incompatible with many polymeric gate insulators for transistor applications. Photolithography may be the alternative choice, but etching solutions and developers often cause damage to polymeric base films and/or gate-insulating layers. In particular, degradation of surface smoothness of gate insulators and/or formation of pinholes are detrimental to organic transistors, because a channel of carrier flow forms at the interface between gate insulators and semiconductors [15].

In this work, we have successfully made via holes though a 900 nm thick polyimide gate-insulating layer, by using a CO_2 laser drilling machine, and fabricated inverter circuits by connecting two high-quality pentacene field-effect transistors on a flexible polyimide base film. Compared with other patterning methods, for example inkjet printing and photolithography, with this method it is possible to keep the surface of the gate insulator clean, smooth, and solvent-free. Thus, no degradation of electrical transport properties of organic transistors has been observed after the laser drilling process. Furthermore, the method is compatible with other polymeric insulators in addition to polyimide and, therefore, we believe that the laser-drilling process is a promising approach to implementing organic transistors for integrated circuits on flexible polymer films.

High-performance organic field-effect transistors with a mobility of ~0.2 $cm^2 V^{-1} s^{-1}$ and an on/off current ratio above 10^5 have been fabricated by a vacuum evaporation process. A cross-sectional illustration of the device is shown schematically in Fig. 1. First, gate electrodes consisting of a 5 nm thick chromium adhesion layer and 100 nm thick gold

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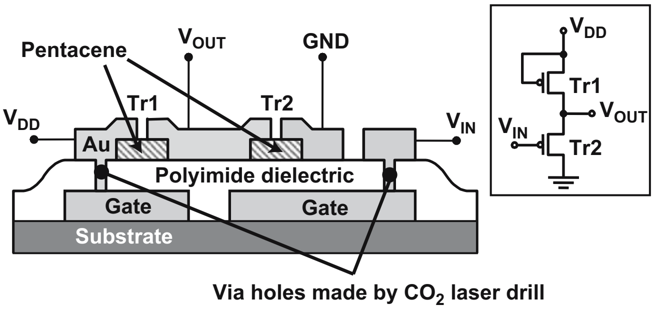

Fig. 1 Cross-sectional illustration of flexible organic transistors on a polyimide base film, and schematic diagram of inverter circuit (*inset*). The thickness of each layer is: polyimide base film 75 μm, gate electrode 100 nm, polyimide gate-insulating layer 900 nm, pentacene 50 nm, and source-drain electrode 40 nm

are deposited on a 75 μm thick polyimide base film (Upilex, Ube Industries) through a shadow mask in a vacuum-evaporation system. A polyimide (Kemtite CT4112; Kyocera Chemical) gate-insulating layer was then prepared by spin coating on a gate electrodes and cured at 180°C for 1 h. Next, via holes are made with a laser drilling machine (ML-G9320; Keyence) which was originally developed as a fine-marking system but which can also function in a machine operation mode. The light source is a CO_2 laser (λ=10.6 μm) with maximum power of 30 W. Laser shots are distributed by a high-speed Galvano scanner controlled by a computer. As shown in Fig. 2, this laser drilling process makes good via holes, diameter of approximately 100 μm, the electrical characteristics of which will be described in detail later. We deposited a 50 nm thick pentacene film by vacuum evaporation and, subsequently, 40 nm thick gold layers, which work as source-drain electrodes and also form contact gold-pads connected to gate electrodes through via holes. Please note that in Fig. 2 the via holes are not filled with metal, and that they have tapered edges. As will be shown later, relatively thin gold layers can lead to good via interconnections with low resistivity. In Fig. 1 the channel length and width of load-transistors (Tr1) are 100 μm and 0.5 mm, respectively, whereas those of switching-transistors (Tr2) are 100 μm and 10 mm, respectively.

Here the output power of the CO_2 laser is changed systematically from 0 to 30 mJ to optimize yields and conductance through the via holes. For this purpose we prepared test structures in which via holes through 900 nm thick polyimide insulating layers are sandwiched between 180 μm square electrodes of 50 nm thick gold, and measured the conductance of each structure.

Similar structures, without via holes, which are relevant to simple capacitors, were also characterized. We found that the number of the structures with 180 μm square electrodes with a leakage current above the noise level (~10 fA at 100 mV) was less than 1%, demonstrating that a pinhole-free gate-insulating layer has been successfully obtained.

Figure 3 shows the dependence on input laser power of the conductance of each via hole. If the incident power is less than approximately 5 mJ, the polyimide film is not removed and, therefore, the measured current is below the noise level. With increasing incident power the area of exposed gold electrodes underneath the polyimide gate-insulating layer increases, and the conductance increases, and saturates at approximately 0.1 Ω^{-1}. When a surplus incident power above 10 mJ is applied some parts of the gold electrodes also start disappearing together with the polyimide gate-insulating layer. It is, however, interesting to note here that fairly good conductance can still be obtained even though the gold electrodes have disappeared or laser shots pass though both polyimide layers and gold electrodes. This is because some parts of the gold electrodes, particularly the side edges, exposed by lasers of excessive power are contacted by top electrodes. As a result, further increasing the incident power still leads to good conductance though the via holes, although it makes the transistors more delicate because much junk is created during exposure to excess laser power.

Yields of via holes are very important in the reliable manufacture of organic circuits. To discuss yields, we counted the number of the devices with conductance exceeding ~$10^{-9}\Omega^{-1}$ of the noise level. Under the best conditions of laser power, thickness of bottom electrodes, and thickness of gate-insulating layer, we confirmed that 99% of devices worked well. Thus, when making three via holes for each pad yields will reach 99.9999%. Further optimization of structural properties and process conditions should improve

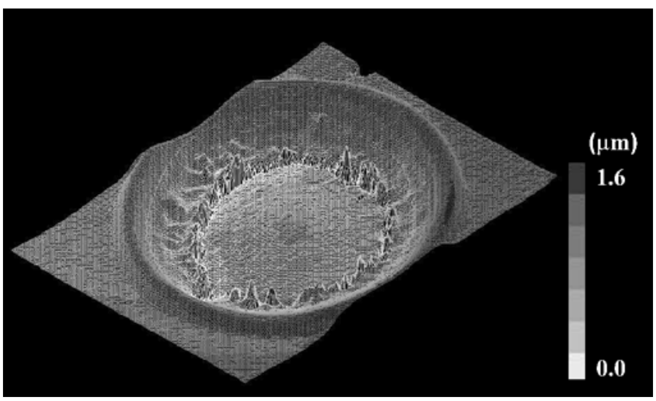

Fig. 2 3D contour of a via hole formed by a laser drilling machine (viewed with a laser microscope)

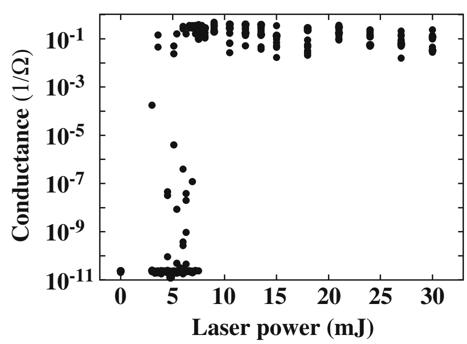

Fig. 3 Dependence of conductance though via holes on input laser power

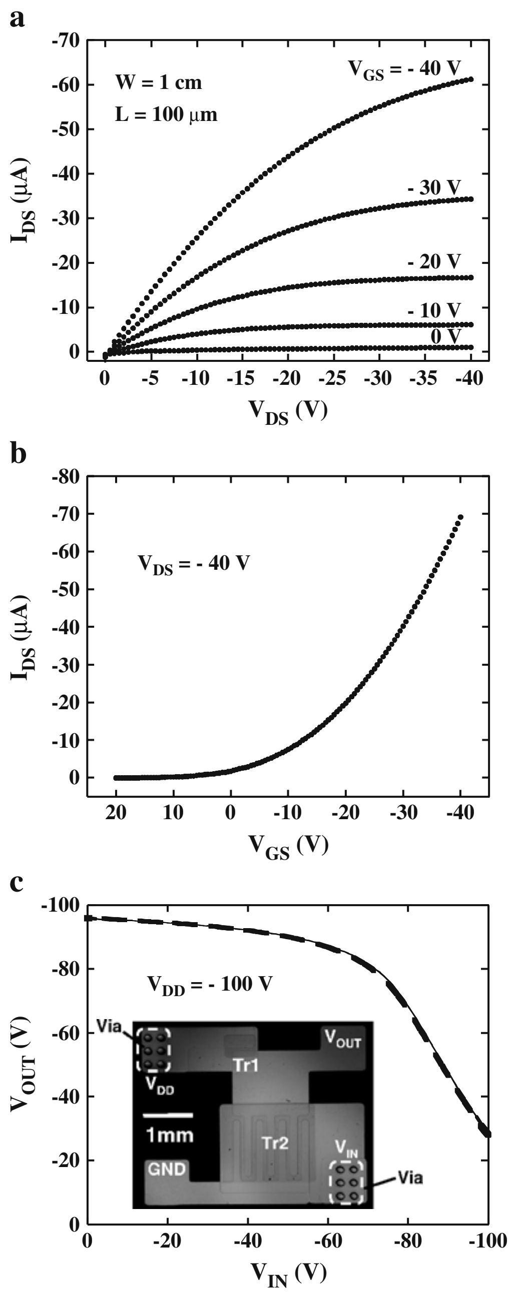

Fig. 4 DC current–voltage characteristics of a single flexible transistor (Tr2, L=100 μm and W=10 mm) (a) as a function of V_{DS} at various V_{GS} and (b) as a function of V_{GS} at V_{DS}=−40 V. (c) Input and output characteristics of a flexible inverter circuit. The *solid line* and *dashed line* indicate the output data when the input was varied from 0 to −100 V and from −100 to 0 V, respectively. A picture of the chip is shown in the *inset*

yields of via holes. Indeed we have found that the stability of the lasers and the thickness of the gold layer underneath polyimide insulators are very important in improving yields. The output stability of the CO_2 laser is ±5%, according to the vendor; to improve stability we warm up the system for more than 1 h before starting experiments. Increasing the thickness of the bottom electrodes also helps to improve yields.

The DC current–voltage characteristics of the organic transistors and the inverter circuits were measured in an ambient environment with a precision semiconductor parameter analyzer (4156C; Agilent Technologies) and a probe station (706f; Micronics Japan). As shown in Fig. 4a, we monitored the source-drain current (I_{DS}) of a single switching-transistor (Tr2) of Fig. 1 as a function of source -drain voltage (V_{DS}). The gate voltage (V_{GS}) was changed from 0 to −40 V in steps of −10 V. The measured mobility and on/off ratio were ~0.2 $cm^2\ V^{-1}\ s^{-1}$ and 10^5, respectively. These values are quite consistent with a similar device manufactured by the same process as mentioned above but without via holes, demonstrating that this laser drilling process does not damage the electrical performance of organic transistors. An electrical transport characteristic of a p-type inverter circuit fabricated on a flexible polyimide film is shown in Fig. 4c. Output voltages (V_{OUT}) were obtained for several input voltages (V_{IN}) from 0 to −100 V (solid line). An experimentally obtained curve obtained with the input voltage reversed from −100 to 0 V is also shown as a dashed line in Fig. 4c. These curves are in excellent agreement, indicating no degradation and thus a reliable inverter circuit.

We would like to emphasize here that this method is a low-cost process which is compatible with large area reel-to-reel manufacture. Indeed, laser drilling machines are now widespread in mass-production of flexible circuit boards, showing their reliability and cost-effective production. Although there is much similarity between the manufacture of flexible circuits boards and of these inverter circuits, two important differences should not be overlooked. First, the thickness of the bottom metal electrodes is much thinner—100 nm in this study whereas it is typically 10 μm on circuit boards. As the thickness of the bottom electrodes is reduced, lasers pass through these also, increasing difficulties. Second, the thickness of polyimide gate-insulating layers for organic circuits is also thinner—900 nm in this study whereas it is at least a few micron for circuit boards. Thin polyimide film does not absorb enough light power, resulting the partial removal of polyimide layers. This study has overcome these two major remaining issues in the optimization of structural properties and process conditions, and thus shows the feasibility of laser drill machining in the fabrication of organic integrated circuits.

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References

1. Baude PF, Ender DA, Haase MA, Kelley TM, Muyres DV, Theiss SD (2003) Appl Phys Lett 82:3964

2. Drury CJ, Mutsaers CMJ, Hart CM, Matters M, de Leeuw DM (1998) Appl Phys Lett 73:108
3. Klauk H, Gundlach DJ, Nichols JA, Jackson TN (1999) IEEE Trans Electron Devices 46:1258
4. Loo Y-L, Someya T, Baldwin KW, Bao Z, Ho P, Dodabalapur A, Katz HE, Rogers JA (2002) Proc Natl Acad Sci USA 99:10252
5. Someya T, Sekitani T, Iba S, Kato Y, Kawaguchi H, Sakurai T (2004) Proc. Natl Acad Sci USA 101:9966
6. Kawaguchi H, Someya T, Sekitani T, Sakurai T (2005) IEEE Journal of Solid-State Circuits 40:177
7. Kane MG, Campi J, Hammond MS, Cuomo FP, Greening B, Sheraw CD, Nichols JA, Gundlach DJ, Huang JR, Kuo CC, Jia L, Klauk H, Jackson TN (2000) IEEE Electron Device Lett 21:534
8. Klauk H, Halik M, Zschieschang U, Schmid G, Radlik W, Weber W (2002) J Appl Phys 92:5259
9. Dimitrakopoulos CD, Malenfant PRL (2002) Adv Mater (Weinheim, Germany) 14:99
10. Klauk H, Gundlach DJ, Jackson TN (1999) IEEE Electron Device Lett 20:289
11. Sirringhaus H, Kawase T, Friend RH, Shimoda T, Inbasekaran M, Wu W, Woo EP (2000) Science 290:2123
12. Kato K, Iba S, Teramoto R, Sekitani T, Someya T, Kawaguchi H, Sakurai T (2004) Appl Phys Lett 84:3789
13. Kawase T, Sirringhaus H, Friend RH, Shimoda T (2001) Proceedings of SPIE, organic field effect transistors, Fichou D, Bao Z, Editors, 4466:80
14. Garnier F, Hajlaoui R, Yassar A, Srivastava P (1994) Science 265:1684
15. Sze S M, (1985) Semiconductor devices, physics and technology (Wiley, New York)