Organic inverter circuits with via holes formed by CO_2 laser drill machine

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Inverter circuits have been made with connecting two high-quality pentacene field-effect transistors. A uniform and pinhole-free 900 nm thick polyimide gate-insulating layer is formed on a flexible polyimide film with gold gate electrodes and partially removed by a CO_2 laser drill machine to make via and contact holes. Subsequent evaporation of a gold layer makes a good electrical connection with a gold gate layer underneath the gate-insulating layer. With optimizing the condition of CO_2 laser drill machine, contact resistance gets to be as low as 3 Ω for 180 μm square electrodes. No degradation of transport properties of organic transistors has been observed before and after the laser drilling process. The present study demonstrates feasibility of laser drilling process to implement organic transistors for integrated circuits on flexible polymer films.

KEYWORDS: organic transistor, laser via process, inverter circuit, pentacene

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Organic field-effect transistors have attracted wide attentions due to their potential application in low-cost large-area flexible electronics, which would be well fitted 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/Vs and on/off ratio of 10^6 have been reported in thin-film transistors of evaporated pentacene.[4,7-11] We have recently achieved much mechanically flexible transistors with mobility of 1 cm^2/Vs using polyimide gate insulators on plastic films.[12] Many of these excellent performances have been demonstrated using discrete organic transistors without device separation.

To implement organic transistors for integrated circuits, it is essential to make patterns of gate insulating layers, particularly, 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] those technologies require precise control of viscosity and careful choice of solvents and, therefore, they are 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, since channel of carrier flows forms at the interface between gate insulators and semiconductors.[15]

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

High-performance organic field-effect transistors with a mobility of ~ 0.2 cm^2/Vs and an on/off current ratio of above 10^5 have been fabricated by a vacuum evaporation process. The cross sectional illustration of the device is schematically shown in Fig. 1. Firstly, gate electrodes, consisting of 5 nm thick chromium adhesion layer and 100 nm thick gold, are deposited on a 75 μm thick polyimide base film (Upilex, Ube Industries Ltd.) through a shadow mask in a vacuum evaporation system. Then, a polyimide (KEMITITE CT4112, Kyocera chemical Co. Ltd.) gate insulating layer was prepared by spin coating on a gate electrodes and cured at 180 ºC for 1hr. Next, via holes are made by a laser drill machine (ML-G9320, Keyence Co. Ltd.), which is originally developed as a fine marking system, but also has a function of 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 drill process makes good via holes with diameter of about 100 μm, whose electrical characteristics will be described in detail later. We deposited by vacuum evaporation a 50 nm thick pentacene film and subsequently 40 nm thick gold layers, which works as source-drain electrodes and also forms contact gold-pads connected to gate electrodes through via holes. In Fig. 1, the channel length and width of load-transistors (Tr1) are 100 μm and 0.5 mm, respectively, while those of switching-transistors (Tr2) are 100 μm and 10 mm, respectively.

Here the output power of a CO_2 laser is changed systematically from 0 to 30 mJ to optimize yields and conductance through via holes. For this purpose, we have made 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 conductance of each structure.

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

Figure 3 shows input laser power dependence of conductance of each via hole. If the incident power is less than about 5 mJ, polyimide film is not removed and therefore measured current is below noise level. With increasing incident power, the area of exposed gold electrodes underneath polyimide gate insulating layer increases and the conductance increases, which saturates around 0.1 Ω^{-1}. When the surplus incident power more than 10 mJ is applied, the some parts of gold electrodes also start disappearing together with polyimide gate-insulating layer. However, it is interesting to note here that fairly good conductance can be still obtained although gold electrodes is disappeared or even laser shots pass though both polyimide layers and gold electrodes. This is because that some parts of gold electrodes, particularly side edges, exposed by lasers with an oversupply are contacted by top electrodes. As a result, the further increase of incident power still lead to good conductance though via holes, while it makes the transistors more delicate due to a lot of junks created during the surplus laser exposing process.

Yields of via holes are very important to manufacture organic circuits reliably. To discuss yields, we counted the number of the devices that showed conductance exceeding $\sim 10^{-9}\,\Omega^{-1}$ of a noise level. Under the best condition of laser power, thickness of bottom electrodes, and thickness of gate-insulating layer, we confirmed that ninety-nine of hundred devices worked well. Thus, making three via holes for each pad, yields will reach 99.9999%. Further optimization of structural parameters and process conditions should improve yields of via holes. Indeed we have found that the stability of lasers and thickness of gold layer underneath polyimide insulators are very important to improve yields. Output stability of CO_2 laser is ±5% according to the vendor's sheet, but to get better stability, we warm up the system for more than 1 hr before starting experiments. Furthermore, increasing the thickness of the bottom electrodes also helps to improve yields.

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

We would like to emphasize here that the present method is a low-cost process, which is compatible with large area reel-to-reel manufacturing. Indeed, laser drill machines are now widely spread in mass-production of flexible circuit boards, showing their reliability and cost effective production. Although there are lots of similarity between manufacturing of flexible circuits boards and that of the present inverter circuits, the following two important differences should not be overlooked. Firstly, the thickness of bottom metal electrodes is extremely thinner, 100 nm in this study, while that is typically 10 μm for circuits board. With decreasing the thickness of bottom electrodes, lasers pass through them as well, increasing the difficulties. Secondly, thickness of polyimide gate-insulating layers is also thinner, 900 nm in the present study, for organic circuits, while that is at least a few μm for circuit boards. Thin polyimide film doesn't absorb enough light power, resulting the partial removal of polyimide layers. The present study has overcome such two major remaining issues with optimizing the structural parameters and process conditions, and thus shows the feasibility of laser drill machining process to fabricate organic integrated circuits.

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References

1. P. F. Baude, D. A. Ender, M. A. Haase, T. W. Kelley, D. V. Muyres and S. D. Theiss: Appl. Phys. Lett. **82** (2003) 3964.
2. C. J. Drury, C. M. J. Mutsaers, C. M. Hart, M. Matters and D. M. de Leeuw: Appl. Phys. Lett. **73** (1998) 108.
3. H. Klauk, D. J. Gundlach, J. A. Nichols and T. N. Jackson: IEEE Trans. Electron Devices **46** (1999) 1258.
4. J. A. Rogers, Y.-L. Loo, T. Someya, K. W. Baldwin, Z. Bao, P. Ho, A. Dodabalapur and H. E. Katz: Proc. Natl. Acad. Sci. USA **99** (2002) 10252.
5. T. Someya, T. Sekitani, S. Iba, Y. Kato, H. Kawaguchi and T. Sakurai: Proc. Natl. Acad. Sci. USA 101 (2004) 9966.
6. H. Kawaguchi, T. Someya, T. Sekitani and T. Sakurai: IEEE Journal of Solid-State Circuits (in press).
7. M. G. Kane, J. Campi, M. S. Hammond, F. P. Cuomo, B. Greening, C. D. Sheraw, J. A. Nichols, D. J. Gundlach, J. R. Huang, C. C. Kuo, L. Jia, H. Klauk and T. N. Jackson: IEEE Electron Device Lett. **21** (2000) 534.
8. H. Klauk, M. Halik, U. Zschieschang, G. Schmid, W. Radlik and W. Weber, J. Appl. Phys. **92** (2002) 5259.
9. C. D. Dimitrakopoulos and P. R. L. Malenfant: Adv. Mater. (Weinheim, Germany) **14** (2002) 99.
10. H. Klauk, D. J. Gundlach and T. N. Jackson: IEEE Electron Device Lett. **20** (1999) 289.
11. H. Sirringhaus, T. Kawase, R. H. Friend, T. Shimoda, M. Inbasekaran, W. Wu and E. P. Woo: Science **290** (2000) 2123.

12. Y. Kato, S. Iba, R. Teramoto, T. Sekitani, T. Someya, H. Kawaguchi and T. Sakurai: Appl. Phys. Lett. **84** (2004) 3789.
13. T. Kawase, H. Sirringhaus, R. H. Friend and T. Shimoda: Proceedings of SPIE, *Organic Field Effect Transistors*, D Fichou, Z Bao, Editors, **4466** (2001) 80.
14. F. Garnier, R. Hajlaoui, A. Yassar and P. Srivastava: Science **265** (1994) 1684.
15. S. M. Sze, *Semiconductor devices, Physics and Technology* (Wiley, New York, 1985)

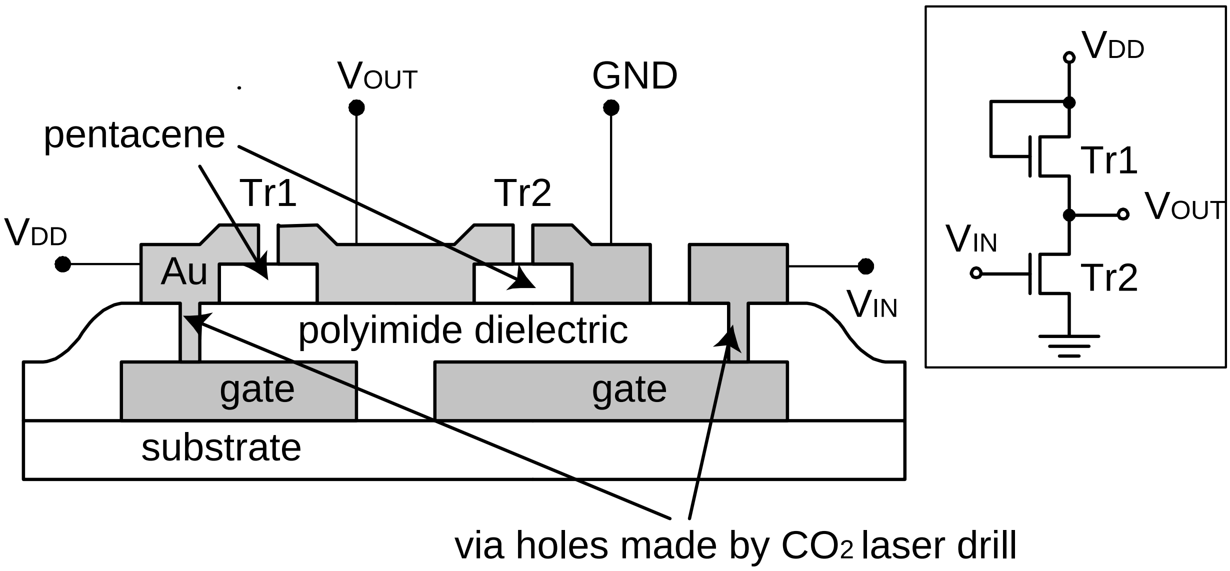
Figure captions

Figure 1: (a) The cross sectional illustration of the flexible organic transistors on a polyimide base film and a schematic inverter circuit (Inset). The thickness of each layer is as follows; a polyimide base film 75 μm, gate electrode 100 nm, polyimide gate insulating layer 900 nm, pentacene 50 nm, and source-drain electrode 40 nm.

Figure 2: 3D contour figure of via hole formed by a laser drill machine, which was taken by a leaser microscope.

Figure 3: Input laser power dependence of conductance though via holes.

Figure 4: (a) DC current-voltage characteristics of a single flexible transistor (Tr2, L = 100 μm and W = 10 mm) as a function of V_{DS} at various V_{GS}, (b) as a function of V_{GS} at V_{DS} = - 40 V. (c) Input and output characteristics of a flexible inverter circuit. Solid line stands for an output data varied input from 0 to -100 V, and dashed line from -100 to 0 V.



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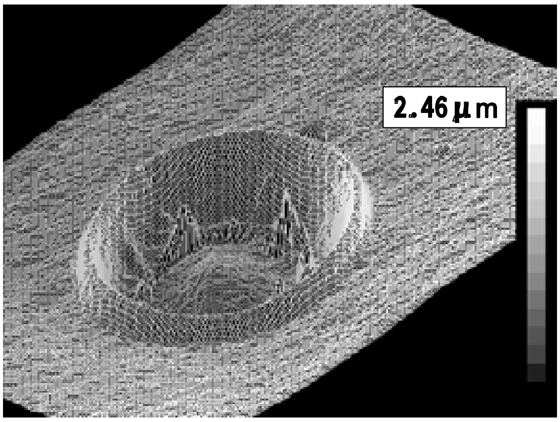

Fig.2 / Iba *et al*.

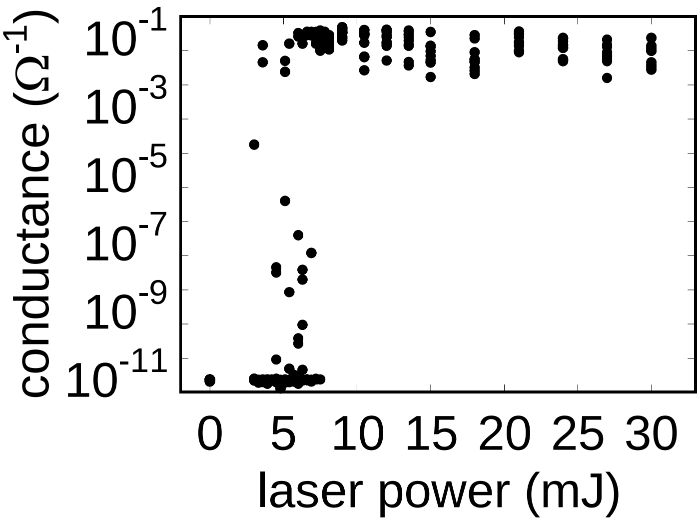

Fig. 3 / Iba et al.

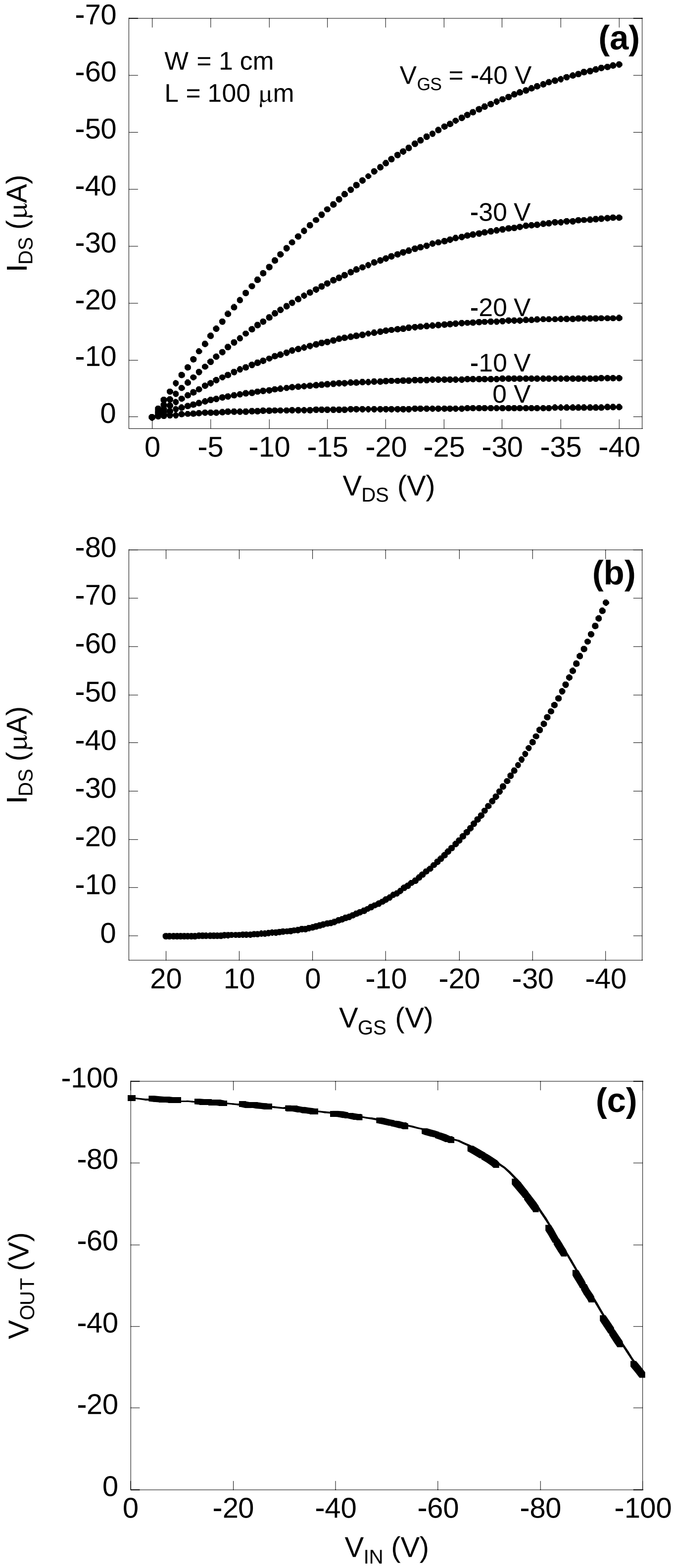

Fig. 4 / Iba *et al.*