Science and applications of organic single-crystal semiconductors

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ABSTRACT

Electronic devices in the future sustainable societies require maximum function with minimum amount of constituent materials and energy cost for production. Electronic functions are often originated from two-dimensional material properties so that device components of large area and small thickness save material consumption. The fundamental electronic functions of both analogue and digital circuits mostly rely on only a nanometer-scale thin layer at the very surface of the semiconductor material due to the very short penetration length of electric field. Recently developed two-dimensional organic single-crystal sheets are highly suited for such future electronics in terms of the minimum material consumption and high electronic performance due to electronic coherence. The presentation focuses on material bases, methods of circuit fabrication, dynamic operation of the devices, and several applications for commercialization.

1 Introduction

Recently developed ultrathin organic semiconductor single crystalline films is suitable for large-area production with low energy consumption; the films are easily formed to large area from solution at relatively low temperature at 80 degrees centigrade [1]. Extremely thin crystal films are controllably grown to a-few molecular layers with the thickness of only 10 nm, so that material cost can be highly limited. Due to a careful material design for restricting harmful molecular vibration, very high carrier mobility more than 10 cm2/Vs is achieved. Furthermore, development of technologies for printed integrated circuits provides a manufacturable process for low-cost platforms for RFID tags and sensing circuitries. Finally, a technology for large-area light-weight display sheets will be demonstrated.

A dynamic transistor model is developed using device parameters such as channel length and overlap length of gate and source electrodes, so that high-frequency transistors are physically designed appropriately including charge-injection resistance [2]. A high cut-off frequency above 45 MHz is actually achieved which are composing elements of organic integrated circuits. Typical integrated circuits of approximately 500 transistors are formed. Moreover, a wireless power-supply is demonstrated using newly developed high-frequency organic rectifiers to operate an RFID tag with the NFC frequency [3].

Such prospect bears increasing reality because of recent research innovations in the field of material chemistry, charge transport physics, and solution processes of printable organic semiconductors. With excellent chemical and thermal stability in recently developed new materials, we are developing simple integrated devices based on CMOS using organic p-type and inorganic n-type FETs. Particularly important are new processing technologies for continuous growth of the organic single-crystalline semiconductor “wafers” from solution and for lithographical patterning of semiconductors and metal electrodes. Successful rectification and identification are demonstrated at 13.56 MHz with printed organic CMOS circuits. Future ten-meter scale large-area display also needs to be more “two-dimensional” to save materials and save energies to carry and equip on billboards.

2 Materials and charge transport physics

A method of continuous crystallization from solution using a meniscus is developed to grow a-few monolayer crystal films to the size of 30 cm x 30 cm [4,5]. The ultrathin organic semiconductor single crystalline films of a p-type organic semiconductor alkyl-naftobenzo-dithiophene (Cn-DNBDT) are easily grown from solution to the wafer scale at temperatures around 80 ^{o}C on a film substrate [5]. With excellent chemical and thermal stability in such materials, integrated devices are developed based on organic CMOS processing technologies. The cut-off frequency of the p-type transistors exceed 75 MHz so that small-scale integrated circuits are practically meaningful. Successful rectification and RF identification are demonstrated at 13.56 MHz with printed organic CMOS circuits [2], so that the organic integrated devices are being commercialized by Pi-Crystal, Inc. to achieve smart society of large-scale multiple-function sensor networks.

Establishing charge coherence and high-mobility charge transport in newly developed solution-crystalized organic transistors is essentially linked to molecular design to achieve band-like charge transport, based on charge coherence in the electronic charge. Such materials show high values of mobility exceeding 10 cm^2/Vs [6].

Furthermore, a two-dimensional hole-gas system is recently induced in the small-molecular assembly with the electronic phases continuously controlled from a wide-gap insulator to an exotic metal with strong electronic correlation [7]. The molecules are weakly bonded to construct organic semiconductor crystals, so that the clean and coherent electronic systems can be formed with relatively small band width (~ 0.5 eV) and on-site Coulomb energy larger than the band width, which is favorable to exhibit exotic electronic phases due to enhanced electronic correlation [8]. With the application of uniaxial strain, the electronic correlation is restricted, resulting in giant strain effect. The mobility exceeds 100 cm^2/Vs at 2 K when 2.8% strain is applied. The coherence is further extended at low temperature, so that Shubnikov-de Haas oscillation is clearly observed at 100 mK with the maximum mobility estimated higher than 700 cm^2/Vs.

3 Applications

We present a scalable flexible active-matrix mini-LED display, which consists of bezelless 3-mm pitch 30 cm x 30 cm independently operating modules. The backplane is based on high-performance organic single-crystal TFTs laminated on screen-printed large-area plastic substrates. The organic TFTs are separately formed on a polyimide film using photolithography. Since their speed and on-current is crucial on flexible films to operate LED chips with sufficient brightness, we employ high-performance organic single-crystal transistors. The value of the mobility exceeds 10 cm^2/Vs. The matrix wirings of silver paint are printed by a large-area screen printing method, so that the vertical and horizontal lines are electrically separated by an insulating layer.

3.1 Large-area flexible signage devices

To meet growing demands of large-area digital signage devices in various situations for advertisement, social events, entertainment and public announcement, more light-weight and low-cost devices are desired than existing passive LED displays. We developed a method to produce LED pixels on printed backplanes of active matrices based on high-mobility organic semiconductors. Due to the light-weight film-based materials of low-cost printed organic active matrices, which is also very effective in reducing power consumption, the present technology meets the requirement for the large-area digital signage.

The present devices are formed on a sheet of PEN film with the thickness of 0.1 mm, so that the large-area movies can be shown on the ceilings or on walls on general buildings without additional construction for reinforcement.

To demonstrate the concept of the large-area LED display sheet, we fabricate modules of 30 cm x 30 cm films with 3-mm pitch active-matrix pixels. The modules are tiled up to any size without bezels, maintaining the same distance between LED chips in the whole multi-module panels.

Fig. 1 An illustration to indicate the usage of the developed flexible mini-LED panels for large-area signage

3.2 Active-matrix driven LED displays

The matrix wirings of silver paint are printed by a large-area screen printing method, so that the vertical and horizontal lines are electrically separated by an insulating layer. The dimensions of the lines are typically 100 μm and the accuracy of the positions is within 50 μm.

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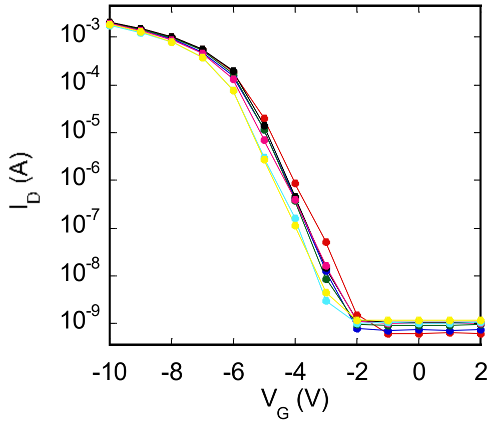

Fig. 2 Transfer characteristics of organic single-crystal TFTs

Figure 2 shows a typical transistor performance of the single crystal organic TFTs, indicating sufficient on-current and well-defined off state. Therefore, the transistor performances of the semiconductors formed from the single-crystals are also sufficiently uniform for the LED pixels.

The active matrix LED panels are formed by laminating the high-performance organic single crystal TFTs and mounting commercially available LED chips on the screen-printed base films. A set of TFTs are incorporated in each unit on a laminated film to illuminate red, green, and blue LEDs, so that the typical circuits are formed in each pixel. A rolling equipment is used to laminate the TFT films in an efficient way. The LED chis with the size of 0.75 mm x 0.75 mm are mounted one by one using a chip mounter.

Finally, outer driving circuits are designed and attached to the edge of the panels, so that any image of 100 x 100 pixels with the RGB colors are transferred to the present flexible LED display panel. The driving circuits are all implemented on a film and attached at the back side of the panel for the bezzelless design of the modules.

Figure 3 shows a connected panel of 1 x 4 modules of the present LED pixels and active-matrix drivers. The luminance level is adjusted for each pixel so that overall pictures are naturally displayed.

Fig. 3 A picture of the scalable flexible mini-LED panel (30 cm x 120 cm)

4 Conclusions

Recent development of organic semiconductors presents significant progress both in small-molecular crystals and semi-crystalline π-conjugated polymers, achieving high carrier mobility based on coherent electronic transport. Extremely thin crystal films of Cn-DNBDT small molecules are successfully grown to a-few molecular layers with the thickness of only 10 nm, so that very high carrier mobility more than 10 cm^2/Vs is achieved to the dimension of 30 cm x 30 cm. The benefit of the ultimately thin and flat platform of mobile charge provides various useful devices when the 2D system makes interfaces to other functional components such as dielectrics or ionic layers and acquire carrier-doping mechanisms. Indeed, current technologies for printed integrated circuits of the organic single-crystal transistors provide a manufacturable process for low-cost platforms for RFID tags, sensing circuitries, and large-area light-weight display sheets.

The method of interfacing electrolytes to the organic semiconductors gives rise to further carrier doping up to 10^{14} cm^{-2}, minimizing the effect of random potentials in the ionic layer. At such carrier concentration, which fills almost 1/8 of the HOMO band, a metallic charge transport with pseudo-gap phase appears as the result of strong electron correlation, reaching carrier mobility of more than 700 cm^2/Vs.

As an application, we demonstrate a novel flexible LED display for large-area signage employing technologies of high-performance solution-processed organic single crystals and an efficient method of laminating TFT chips to mount on the all pixels. The fabrication is on the way for mass-production, so that products such as 10-m size display sheets hung from the ceiling appear in the market in near future.

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