Studies of pentacene-based thin film devices produced by cluster beam deposition methods

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Abstract

The cluster beam deposition (CBD) method has been applied to fabricate and characterize pentacene-based thin film devices. Atomic force microscopy, scanning electron microscopy, and X-ray diffraction measurements demonstrate that the weakly bound and highly directional cluster beam is effective in producing a highly ordered structure close to a single crystal with a uniform flat film surface. The hole carrier mobility has been determined to be approximately $10^{-4}$ cm²/Vs as a lower bound from the $J-V$ characteristics for the ITO/pentacene/Al devices. The Schottky barrier-type MOSFETs with a 10 μm long channel length have been produced and show a typical source–drain current modulation behavior with different gate voltage.

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1. Introduction

Thin film field-effect transistors utilizing organic semiconductors have attracted much attention due to their potential applications in low-cost integrated circuits and future flat panel displays. Most of studies have been directed toward improving characteristics such as field-effect mobility and on–off current ratio by developing and optimizing new organic materials. The performance of some organic field-effect transistors (OFETs) now stands in the state of the competition with amorphous silicon FETs in recent years. This is exemplified by pentacene (C₁₄H₂₂) which consists of five aligned condensed benzene rings. Pentacene films with an ordered morphology have been obtained using vacuum evaporation, pulsed laser deposition or chemically vapor deposition [1,2]. In a case the high charge carrier mobility up to ~1 cm²/Vs has been reported, which is believed to be due to the single crystallinity of the organic films [3]. However, such a high value is known to be reproducible in very narrow ranges of the particular deposition parameters.
In this study, we present the fabrication and characterization of the pentacene-based thin film devices using cluster beam deposition (CBD) methods. The unique advantages of employing cluster beam are the high directionality and translational kinetic energy of the beam when the gas molecules undergo adiabatic expansion into high vacuum. Since the clusters are composed of weakly bound molecules, the collision of clusters with the substrate of interest induces facile fragmentation into individual molecules and the subsequent active surface migration leads to significant improvement in surface morphology, crystalline quality, film packing density and low substrate-temperature deposition that cannot be achieved by other conventional evaporation methods.

2. Experimental

Pentacene was deposited on the substrate by the home-made CBD apparatus recently built. The system employed in this work will be described in detail elsewhere, and here only a brief relevant account is presented [4]. The schematic diagram of the apparatus is shown in Fig. 1. The apparatus consists of the evaporation crucible cell, the ionization and extraction electrodes, the drift region and the substrate. The chamber was pumped by a 10 in. baffled diffusion pump and the average pressure was maintained below $1 \times 10^{-6}$ Torr. The source material pentacene placed inside the enclosed cylindrical cell with a nozzle (1.0-mm diameter, 1.0-mm long) was evaporated by the resistive heating of the cell at 600 K. As the sublimated gas molecules expanded into a high vacuum region, the pentacene sample underwent adiabatic expansion, leading to the formation of weakly bound neutral clusters. In this study the pentacene layer was deposited using a two-step deposition scheme. First, the neutral pentacene clusters were directly deposited onto the substrate as a buffer layer. Afterward, some fraction of the neutral clusters underwent ionization by the electron impact source and the resultant ionized cluster beam passed through an extraction electrode, where the beam was accelerated to travel the drift region and then deposited onto the buffer-layer deposited substrate. The two-step deposition has extensively been employed in the epitaxial growth to improve crystalline quality and particularly to avoid direct ion bombardment in this study. Normally several substrates were simultaneously deposited at room temperature throughout the deposition process. The growth rate of the film was dominated by the temperature of the evaporation crucible cell and was measured to be typically 0.3 A/s by a thickness monitor. The film thickness, crystallinity, surface and lateral morphology were measured by an alpha step surface profile monitor, X-ray diffraction (XRD), atomic force microscopy (AFM), and scanning electron microscopy (SEM). Device characteristics such as current or current density vs. voltage ($I-V$, $J-V$) were also obtained and analyzed.

3. Results and discussion

3.1. Characterization of pentacene films

The surface of the pentacene thin films has been examined by recording AFM images. Fig. 2 shows the two-dimensional micrograph for the pentacene films around 6000 Å thickness deposited on glass substrates and demonstrates grain crystallites ranging from 0.3 to 0.6 μm in diameter. The quantitative values of the root-mean-square roughness ($R_{\text{rms}}$) were obtained through conducting section analysis and were estimated to be
typically 40 Å. Such a low $R_{\text{rms}}$ value compared to the film thickness suggests that the cluster beam employed in this work leads to extremely smooth and uniform thin films.

Fig. 3(a) shows the XRD patterns of the films deposited on glass substrates. These films showed a highly ordered structure with three distinctive multiple peaks with a 002 plane spacing $d_{002} \approx 7.88$ Å. The single crystal of pentacene is reported to have a triclinic structure with $d_{001} \approx 14.5$ Å [5]. A broad maximum at $2\theta$ of around 20–25° comes from the glass substrate. To increase crystalline quality a buffer layer using hexamethyldisilazane (HMDS) was first spin-coated on the glass substrate surface, and afterward the pentacene was deposited via the CBD method. The HMDS is an amphiphilic surfactant molecule capable of forming bonds with hydrophobic pentacene and hydrophilic SiO$_2$ simultaneously, leading to the reduction of the lattice mismatch. Fig. 3(b) demonstrates improved crystallinity and more multiple peaks with $d_{001}$ being equal to 14.3 Å closer to that of the single crystal.

### 3.2. Hole carrier mobility in pentacene films

To understand the carrier transport properties of the pentacene films, studies of the $J$–$V$ characteristics were performed. On top of the optically transparent indium tin oxide (ITO) as an anode, the pentacene film was deposited to the thickness of 1500 Å using the CBD method and then vacuum-evaporated Al was superimposed as a cathode. The $J$–$V$ curves with schematic energy level diagram for the device are shown in Fig. 4. With increasing the bias voltages the current density clearly shows a series of successive transitions in the conduction mechanisms. The slope of the $J$–$V$ curve can be fitted with theoretical models to obtain several important quantities related to the transport. The intermediate region corresponding to the space-charge limited (SCL) case can be fitted with a power law ($J \propto V^m$) with exponent 2 and the hole carrier mobility is determined by the following so-called modified Mott–Gurney equation,

$$J_{\text{SCL}} = \frac{9}{8} \varepsilon_0 \mu_{\text{eff}} V^2 / d^3$$

where $\mu_{\text{eff}}$ is the effective carrier mobility, $d$ the thickness of the device and $\varepsilon$ the dielectric constant.
(ε = 4 for pentacene) [6]. The carrier mobility was calculated to be about 10^{-4} cm^2/Vs. At the threshold voltage (V_{th} = 5.6 V) separating the SCL regime from the trapped-charge limited regime, an abrupt transition is observed and the trap concentration can be estimated to be ≈ 1.2 \times 10^{17} cm^{-3} [7]. The results suggest that while a single crystalline structure with a smooth surface is obtained using the CBD method, the cluster beam induces a larger number of trap sites in the grain-shaped microcrystallites. Therefore, the obtained mobility as a lower bound is expected to increase with improving the crystalline quality, which is now currently under investigation [8].

3.3. Fabrication and characterization of MOSFETs

The Schottky barrier-type metal oxide semiconductor FETs (MOSFETs) have been fabricated. Fig. 5 shows the cross-sectional SEM photograph and the schematic diagram of the device with a well-defined film surface. A highly doped n-type silicon gate electrode covered with thermally grown 100 nm thick SiO$_2$ was used as a substrate and aluminum source and drain contacts with separation length (L) of 10 μm and channel width (W) of 400 μm were superimposed. Pentacene was deposited to the thickness of 6000 Å using the CBD method. Fig. 5(c) shows the typical source–drain current modulation behavior with different gate voltages for p-type MOSFETs. From the fits of the $I–V$ characteristics the hole carrier mobility and on/off current ratio are estimated to be about $10^{-6}$ cm$^2$/Vs, and $10^2$–$10^3$, respectively. Since the contact between Al source and pentacene layers was not ohmic, the carrier injection was found to be low and showed the behavior limited by the tunnelling through the Schottky barrier. It is expected that the current will increase sharply as the temperature ($T$) increases or the barrier height ($\phi$) decreases from the following Schottky emission equation:

$$I \propto T^2 \exp \left(-\frac{q(\phi - \sqrt{qE/4\pi\varepsilon})}{kT}\right)$$

The current was measured at two different temperature 330 and 360 K, and increased by about a factor of 70 (about 3.2–225 pA), which is consistent with the emission theory. The HMDS buffer was also used between SiO$_2$ and pentacene, and the mobility and the source–drain current increased by about a factor of 2 orders (about $10^{-6}$–$10^{-4}$ cm$^2$/Vs) due to the higher crystalline quality.

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