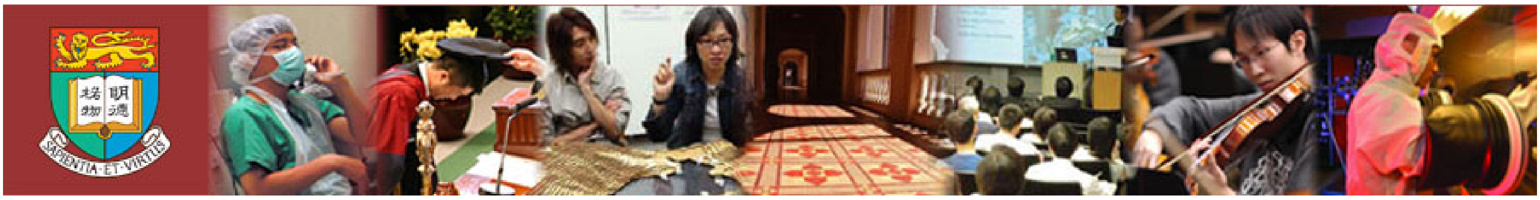

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High-Performance Pentacene Thin-Film Transistor With High-κ HfLaON as Gate Dielectric

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***Abstract*— Pentacene organic thin-film transistor (OTFT) using high-*k* HfLaON gate dielectric is proposed, and the effects of varying its nitrogen content are studied. The HfLaON film is deposited using reactive sputtering of Hf–La target in Ar/O_2/N_2 ambience with different N_2 flow rates and then annealed in N_2. All the OTFTs can operate at low voltage with a threshold voltage as low as −0.53 V. The OTFT with an optimal nitrogen content can achieve a carrier mobility of 0.71 $cm^2/V\cdot s$, which is about twice that of its counterpart with HfLaO gate dielectric.**

***Index Terms*— Fluorination, HfLaON, high-κ dielectric, organic thin-film transistor (OTFT).**

I. INTRODUCTION

ORGANIC thin-film transistors (OTFTs) have been intensively studied due to their potential applications in radio-frequency (RF) identification tags, sensors, and large-area flexible displays [1]. Over the last four decades, various organic semiconductor materials have been proposed to fabricate the OTFTs. Among them, pentacene OTFTs can exhibit a high carrier mobility (>1 cm^2/V s), comparable with that of amorphous-silicon TFT [2]. However, pentacene OTFTs fabricated with SiO_2 gate dielectric operate at high supply voltage with high threshold voltage (>5 V) [3], thus requiring special power source or converter circuit in applications. High-κ dielectric material is essential to gain high carrier mobility and low threshold voltage [4]. Lanthanum incorporated in HfO_2 could increase the carrier mobility of the OTFTs [5]. In addition, annealing HfLaO in NH_3 could improve the dielectric property through nitrogen incorporation, and thus enhanced performance was achieved for the OTFTs [6]. Therefore, in this letter, HfLaON films with different nitrogen contents are prepared by RF sputtering, and then OTFTs are fabricated with the HfLaON films as gate dielectric. With optimal nitrogen incorporation, the electrical properties of the OTFT can be greatly improved. Furthermore, the low-frequency noise (LFN) spectrum of the devices is used to study the defects located at the HfLaON/pentacene interface due to its high sensitivity to carrier-number fluctuation in the devices [7].

II. EXPERIMENTAL DETAILS

Initially, silicon wafers (n-type, <100>, resistivity of 0.5–1 Ω· cm) were cleaned according to the standard RCA method and dipped in 2% HF acid to remove the native oxide. Then, HfLaON films were deposited by sputtering in an Ar/O_2/N_2 ambience with different N_2 flow rates: 1) 0 sccm for sample A; 2) 3 sccm for sample B; 3) 6 sccm for sample C; and 4) 12 sccm for sample D. A RF sputterer (Denton Vacuum LLC Discovery 635) and Hf–La metal target (with 40% La) were employed to prepare the HfLaON films at a RF power of 40 W. The flow rate of Ar/O_2 was kept at 24:3 sccm for all the sputtering conditions. Then, the samples were annealed in N_2 at 400 °C. Next, 30-nm pentacene (99%, purchased from Sigma-Aldrich without purification) was evaporated on the dielectrics by an evaporator (Edwards Auto 306) in high vacuum (4×10^{-6} torr) at a deposition rate of 1 nm/min, monitored by a quartz-crystal oscillator. Finally, gold was deposited on the 30-nm pentacene film by evaporation through a shadow mask to form the drain and source electrodes of the OTFTs. The width and length of the channel on the shadow mask were 200 and 30 μm, respectively.

The I–V characteristics and LFN of the transistors were measured by HP 4145B semiconductor parameter analyzer, a Berkeley Technology Associates FET Noise Analyzer Model 9603 and HP 35665A Dynamic Signal Analyzer. An Al/HfLaON /heavily-doped Si structure was also fabricated by lithography for measuring the dielectric capacitance with HP 4284A precision LCR meter. The thickness of the dielectric films was measured by a Wvase 32 ellipsometer. A Nanopics 2100 atomic force microscopy was employed to record the surface morphology of the dielectrics. All the measurements were conducted at room temperature, in air, and under shielded environment.

III. RESULTS AND DISCUSSION

All the OTFTs show good field-effect property and saturation behavior, and they can operate at a very low voltage <5 V as shown in Fig. 1. The drain current of the sample C is the largest among the devices and is about twice than that of the control sample A without nitrogen incorporation.

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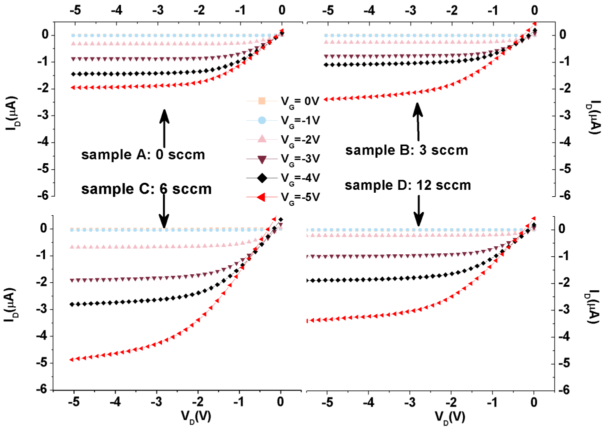

Fig. 1. Output characteristics of the OTFTs with different N_2 flow rates in sputtering.

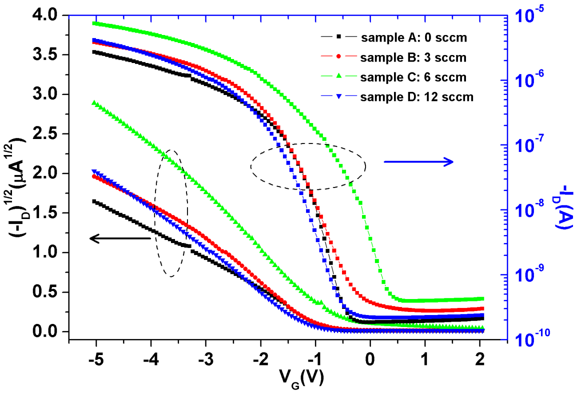

Fig. 2. Transfer characteristics of the OTFTs at $V_D = -5$ V with different N_2 flow rates in sputtering.

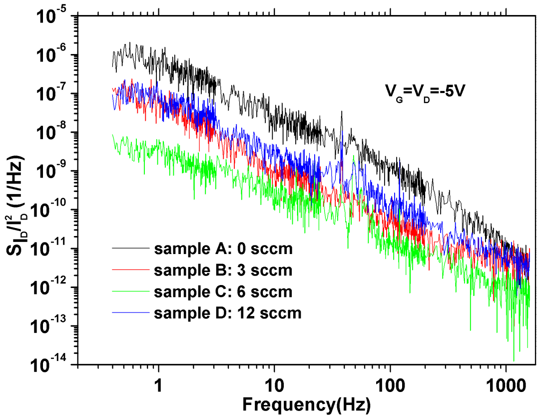

Fig. 3. LFN characteristics of OTFTs with different N_2 flow rates during sputtering at $V_G = V_D = -5$ V.

TABLE I

PARAMETERS OF THE OTFTS WITH ANNEALING IN N_2 AT 400 °C

Sample No.	A	B	C	D
Nitrogen Flow Rate (sccm)	0	3	6	12
Average Carrier Mobility (cm^2/V·s)(Highest)	0.312 (0.393)	0.341 (0.454)	0.578 (0.711)	0.330 (0.419)
Threshold Voltage (V)	-0.587	-0.529	-0.696	-0.722
SS (V/dec)	0.572	0.517	0.427	0.566
On/Off Current Ratio (10^3)	18.3	13.3	19.9	17.5
I_D @ $V_D = V_G = -5$ V (μA)	3.94	4.66	8.13	5.12
C_{ox} (μF/cm^2)	0.240	0.279	0.297	0.294
t_{ox} (nm)	39.9	37.0	34.2	32.9
κ	10.8	11.6	11.5	10.9
μC_{ox} (μF/ V·s)	0.075	0.095	0.172	0.097
RMS Roughness (nm)	0.137	0.159	0.138	0.142
N_t (×10^{19} $eV^{-1}cm^{-3}$)	45.9	7.07	0.263	0.302

The transfer characteristics of the OTFTs are shown in Fig. 2. According to the conventional current–voltage expression of metal-oxide-semiconductor field-effect transistor (MOSFET) operating in saturation region, the carrier mobility (μ) can be extracted by

$$\mu = \frac{2L\left(\frac{\partial\sqrt{I_D}}{\partial V_G}\right)^2}{\mathrm{WC_{ox}}} \tag{1}$$

where W is the channel width, L is the channel length, C_{ox} is the insulator capacitance per unit area, I_D is the drain current, V_G is the gate voltage, and V_T is the threshold voltage.

For OTFT based on high-κ dielectric, the fluctuation in carrier number caused by the trapping and detrapping processes in the gate dielectric dominates the fluctuation in the drain current. Therefore, the power spectral density of the drain current $S_{I_D}(f)$ can be expressed as follows [8]:

$$\frac{S_{I_D}(f)}{I_D^2} = \frac{g_m^2}{I_D^2}\frac{q^2kTN_t}{\mathrm{WLC_{ox}^2}f^{\gamma}} \tag{2}$$

where q is the electron charge, k is Boltzmann constant, T is the temperature (300 K), f is the frequency, and g_m is the transconductance.

From (2), the trap density (N_t) can be calculated when the power spectral density of drain current at a frequency of 1 Hz is measured. The LFN spectrum is measured and shown in Fig. 3. The normalized noise of the sample C exhibits the lowest normalized spectral density, indicating that the bulk trap density is the lowest among the devices, because the traps in the bulk and at the surface of the dielectric can be passivated by nitrogen atoms incorporated during the sputtering process.

The key parameters of the OTFTs are extracted and listed in Table I. The threshold voltages of all the samples are less than −0.8 V. The average carrier mobility of the sample C is 0.578 cm^2/V s, the highest among the samples. The carrier mobility increases with the nitrogen flow rate in the sputtering process but then decreases for flow rate >6 sccm. Correspondingly, the trap density extracted from the LFN spectrum decreases with increasing flow rate and reaches the lowest at a flow rate of 6 sccm. Further increasing the flow rate increases the trap density as more nitrogen atoms are incorporated in the film during the sputtering. In the dielectric and at the interface, there are two types of traps: 1) acceptorlike traps and 2) donorlike traps. The onset voltage of the sample C is the highest, indicating that the nitrogen incorporation can create acceptorlike traps in the gate dielectric [9]. However, the carrier mobility of the holes in the conduction channel is not affected by these acceptorlike traps [10]. On the other hand, the nitrogen incorporation can passivate the donorlike traps in the bulk and at the surface of the gate oxide, resulting in less

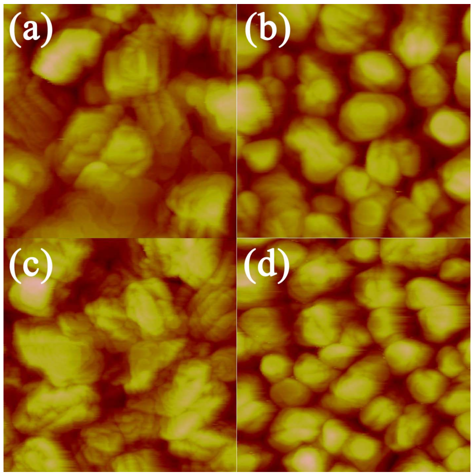

Fig. 4. AFM image of pentacene film grown on HfLaON gate dielectrics with different N_2 flow rates in sputtering. (a) 0 sccm N_2. (b) 3 sccm N_2. (c) 6 sccm N_2. (d) 12 sccm N_2. The area is $1 \times 1\ \mu m^2$.

carrier scattering and thus higher carrier mobility. Compared with the sample C, excessive nitrogen incorporation in the sample D can create too much positive fixed oxide charge [11], which results in stronger carrier scattering and thus lower carrier mobility.

The morphologies of pentacene and gate dielectric are characterized by AFM. Rougher dielectric surface results in the growth of smaller pentacene grains [12], [13]. The samples A and C have smaller rms roughness (i.e., smoother gate dielectric surface) as listed in Table I and thus larger pentacene grains as shown in Fig. 4. However, the carrier mobility of the sample C is much higher than that of the sample A. One explanation is that with the passivation effect of nitrogen incorporation, the sample C has much less traps (Table I) for scattering the carriers. Therefore, pentacene grain size, interface-trap density, and dielectric surface roughness all have significant effect on the carrier mobility of OTFTs. Larger grain size, lower trap density, and smaller surface roughness can result in higher carrier mobility as demonstrated by the sample C.

IV. CONCLUSION

Pentacene OTFT with high-*k* HfLaON as gate dielectric is proposed. The effects of varying the nitrogen content during sputtering and annealing the HfLaON film in N_2 are investigated. With proper nitrogen incorporation, the carrier mobility of the OTFT can be greatly improved because nitrogen atoms can passivate the traps in the oxide bulk and at the HfLaON/pentacene interface. However, too much nitrogen incorporation results in rougher oxide surface and smaller grain size, thus degrading the electrical properties of the OTFT. The LFN measurement verifies that appropriate nitrogen incorporation can reduce the trap density. In addition, AFM supports that larger pentacene grain and smoother dielectric surface induced by nitrogen incorporation can contribute to higher carrier mobility. This letter demonstrates that HfLaO film with proper nitrogen incorporation can act as the gate dielectric of high-mobility low-voltage OTFTs.

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