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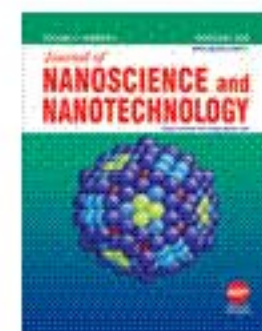
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## **S** Low-Voltage-Driven Pentacene Thin-Film Transistors with Cross-Linked Poly(4-vinylphenol)/High- $k$ $\text{Bi}_5\text{Nb}_3\text{O}_{15}$ Hybrid Dielectric for Phototransistor

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### Abstract:

This paper describes the fabrication of pentacene thin-film transistors (TFTs) with an organic/inorganic hybrid gate dielectric, consisting of cross-linked poly(4-vinylphenol) (PVP) and  $\text{Bi}_5\text{Nb}_3\text{O}_{15}$ . A 300-nm-thick  $\text{Bi}_5\text{Nb}_3\text{O}_{15}$  dielectric film, grown at room temperature, exhibits a high dielectric constant (high- $\epsilon_c$ ) value of 40 but has an undesirable interface with organic semiconductors (OSC). To form better interfaces with OSC, a cross-linked PVP dielectric was stacked on the  $\text{Bi}_5\text{Nb}_3\text{O}_{15}$  dielectric. It is shown that, with the introduction of a hybrid dielectric, our devices not only can be operated at a low voltage ( $\sim -5$  V) but also have improved electrical characteristics and photoresponse, including a field-effect mobility of  $0.72 \text{ cm}^2/\text{Vs}$ , current sub-threshold slopes of  $0.29 \text{ V/decade}$ , and a photoresponse of 4.84 at a gate bias  $V_G = 0 \text{ V}$  under  $100 \text{ mW/cm}^2$  AM 1.5 illumination.

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# Low-Voltage-Driven Pentacene Thin-Film Transistors with Cross-Linked Poly(4-vinylphenol)/High- $k$ Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> Hybrid Dielectric for Phototransistor

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This paper describes the fabrication of pentacene thin-film transistors (TFTs) with an organic/ inorganic hybrid gate dielectric, consisting of cross-linked poly(4-vinylphenol) (PVP) and Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub>. A 300-nm-thick Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric film, grown at room temperature, exhibits a high dielectric constant (high- $k$ ) value of 40 but has an undesirable interface with organic semiconductors (OSC). To form better interfaces with OSC, a cross-linked PVP dielectric was stacked on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric. It is shown that, with the introduction of a hybrid dielectric, our devices not only can be operated at a low voltage ( $\sim -5$  V) but also have improved electrical characteristics and photoresponse, including a field-effect mobility of 0.72 cm<sup>2</sup>/V·s, current sub-threshold slopes of 0.29 V/decade, and a photoresponse of 4.84 at a gate bias  $V_G = 0$  V under 100 mW/cm<sup>2</sup> AM 1.5 illumination.

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**Keywords:** Organic Thin Film Transistor, Low Voltage, Hybrid Gate Dielectric, Pentacene, Poly(4-vinylphenol), Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub>, Photoresponse.

## 1. INTRODUCTION

In recent years impressive progress has been made in the study of organic thin-film transistors (OTFTs), with the transistors being put to work in such applications as large-area organic electronics and displays.<sup>1–3</sup> Recently, OTFTs have also been considered as candidates for application to optoelectronics, so several research groups have reported on the properties of photoresponsive OTFTs based on conjugated polymer/fullerene blends, bifunctional spiro compounds, and  $p$ -type organic semiconductors.<sup>4–6</sup> A disadvantage of such OTFTs is that they require a high operating voltage ( $>20$  V) compared to conventional inorganic TFTs and thus are expected to be of limited use in portable applications because of their high power consumption. To avoid this issue, one must reduce the operating voltage and thus reduce the power consumption, and this in turn depends on the selection of the dielectric materials used in the transistor as well as their thickness. For a low-voltage operating OTFT, the gate dielectric should have a high capacitance value, which can be achieved by

employing a high- $k$  dielectric material or else by reducing the dielectric thickness. Since reports of the first use of high- $k$  oxide dielectrics to reduce the operating voltage of OTFTs, many high- $k$  oxide dielectrics, such as Ta<sub>2</sub>O<sub>5</sub> and TiO<sub>2</sub>, have received attention for possible use as gate dielectrics.<sup>7–8</sup> However, not only must these high- $k$  dielectric materials be thick in order to avoid leakage, but they also form undesirable interfaces with the organic semiconductor (OSC) film because of their hydrophilic surfaces. One way to avoid the formation of these undesirable surfaces is to insert an organic dielectric between the inorganic dielectric and the OSC film, which can form an interface that interacts with the OSC layer, which in turn smoothes the surface roughness affecting the ordering of OSC molecules during active channel layer formation and thus enhancing the field effect mobility.<sup>9</sup> By stacking the organic dielectric on the inorganic dielectric, such OTFTs combine the advantages of an organic dielectric's good surface conditions with the low-voltage-driven gate made from an inorganic dielectric. Thus a certain amount of research has been done on the fabrication of low-voltage-driven OTFTs with organic and high- $k$  inorganic hybrid dielectrics.<sup>10–12</sup> Unfortunately, however, those devices tend

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to have relatively high leakage levels and also to have inferior sub-threshold slope characteristics. In this paper, we describe the development of pentacene TFTs with cross-linked poly(4-vinylphenol) (PVP)/high-*k* Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> hybrid gate dielectrics, with which we explored some of these issues. Additionally, the photoresponsive characteristics of these materials were investigated. A Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric film deposited at room temperature has a high-*k* value of 40 and a relatively low leakage current density.<sup>13</sup> Moreover, a stacked PVP dielectric can improve the condition of the OSC/dielectric interface. Consequently, the proposed hybrid dielectric could play a role as a superior dielectric layer for low-voltage-driven OTFTs.

## 2. EXPERIMENTAL DETAILS

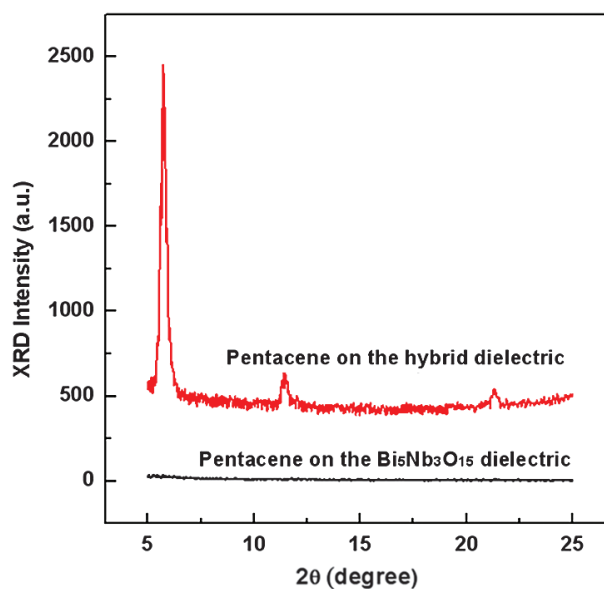
The substrate and gate electrode are indium-tin-oxide (ITO) glass (sheet resistance  $\sim 10.0$  ohm/sq), and a 300-nm-thick Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> film is grown on the 100-nm-thick ITO glass substrate with RF magnetron sputtering, using a 3-inch-diameter Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> target. The film deposition is carried out at room temperature in a mixed oxygen and argon (O<sub>2</sub>:Ar = 1:4) atmosphere at a total pressure of 8.5 mTorr. A cross-linked PVP film is then deposited on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> thin film by spin-coating from a solution of PVP, and the film is cured at 200 °C for five minutes. The PVP solution is prepared by mixing PVP powder (Sigma-Aldrich, Mw  $\sim 20,000$ ) with propylene glycol monomethyl ether acetate (PGMEA) and poly(melamine-co-formaldehyde) (Sigma-Aldrich, Mw  $\sim 511$ ) added as a cross-linking agent. After the hybrid dielectric is formed, a 70-nm-thick pentacene active layer is deposited by a thermal evaporator (DOV Co., Ltd.) with a deposition rate of 0.4 Å/s for the active layer. Then a 250-nm-thick source and drain electrodes made of Au are deposited using the thermal evaporator; the device's channel length and width are 100  $\mu\text{m}$  and 2500  $\mu\text{m}$ . The surface morphology and the structural property of the proposed devices were studied with atomic force microscopy (AFM, Dimension 3100) and X-ray diffraction (XRD, D/max 2200V) spectroscopy in the symmetric reflection coupled  $\theta$ - $2\theta$  arrangement with a CuK $\alpha_1$  radiation ( $\lambda_{\text{K}\alpha_1} = 1.54$  Å) X-ray source. The electrical characteristics of the pentacene TFTs were measured using a semiconductor characterization system (Keithley SCS 4200) in a dark box.

## 3. RESULTS AND DISCUSSION

To evaluate the dielectric characteristics of the proposed dielectric materials, we fabricated the metal-insulator-metal (MIM) structure and measured the electrical characteristics at 100 kHz. The capacitance density of Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> (300 nm) and of the PVP (70 nm)/Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> (300 nm) hybrid dielectric are 116 nF/cm<sup>2</sup> and 34.4 nF/cm<sup>2</sup>, respectively. The hybrid dielectric has a smaller capacitance

because of the series connection of the PVP layer. The results imply an effective dielectric constant ( $k_{\text{eff}}$ ) value of 14.8 after the PVP dielectric is stacked on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric; this  $k_{\text{eff}}$  is larger than the effective dielectric constant of either Al<sub>2</sub>O<sub>3</sub> ( $k_{\text{bulk}} = 8.4$ ) or Si<sub>3</sub>N<sub>4</sub> ( $k_{\text{bulk}} = 6.2$ ).<sup>8</sup> To verify the dielectric properties of the proposed hybrid dielectrics, we measured the current density–electric field ( $J$ - $E$ ) characteristics with a programmable electrometer (Keithley 617). The leakage current density of a single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> is relatively low, but its dielectric strength ( $< 0.4$  MV/cm) makes it insufficient for use as the dielectric layer. By stacking the PVP layer on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> hybrid dielectric, we can create a reasonably effective dielectric, which exhibits a leakage current of  $\sim 10^{-7}$  A/cm<sup>2</sup> at a dielectric strength of  $\sim 1.6$  MV/cm (not shown here). In order to study the surface morphology, we compared the crystallization and grain boundaries of the pentacene films on two different dielectrics.

Figure 1 shows the surface conditions of the pentacene films as revealed by XRD spectra. The upper spectra exhibit a series of (0 0 *l*) diffraction peaks, indicating that the pentacene film on the hybrid dielectric has a well-organized molecular structure. As shown in Figure 1, the maximum peak is observed at 5.72°, corresponding to a layer-by-layer separation of 17.2 Å, which is in very close agreement with the thin film phase reported in the literature.<sup>14</sup> By contrast, the lower spectra for the pentacene film on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric exhibits negligible peaks, which implies amorphous or disorderly grown pentacene molecules, confirmed by the AFM images. As shown in the figures, the pentacene film on the hybrid dielectric has a larger average grain size than the pentacene film on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> film. Generally, having more



**Fig. 1.** (a) XRD spectra of pentacene films on the hybrid dielectric (PVP/Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub>) and high-*k* dielectric (Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub>).

grain boundaries leads to more potential barriers, which in turn limits the distance that a charge can move without running into an obstacle; thus, a larger grain size is advantageous for the movement of the charges.<sup>15</sup>

Furthermore, the well-organized pentacene films on the hybrid dielectric may enhance the field effect mobility of the device because of the better  $\pi$ -orbital overlap among the neighboring pentacene molecules.<sup>16</sup> Clearly, the PVP dielectric has a positive effect on the crystallization of the pentacene molecules and increases the grain size, which may improve the electrical performance of pentacene TFTs. Figures 2(a and b) and their insets show the transfer characteristics ( $\log(-I_D)$  vs.  $V_G$  and  $\sqrt{-I_D}$  vs.  $V_G$ ) and the output characteristics ( $I_D$  vs.  $V_D$ ) of the proposed pentacene TFTs with the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric and the hybrid dielectric, respectively. The pentacene TFT with the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric exhibits an on-state current ( $I_{on}$ ) of 0.18  $\mu$ A, a current on/off ratio of  $2 \times 10^3$ ,

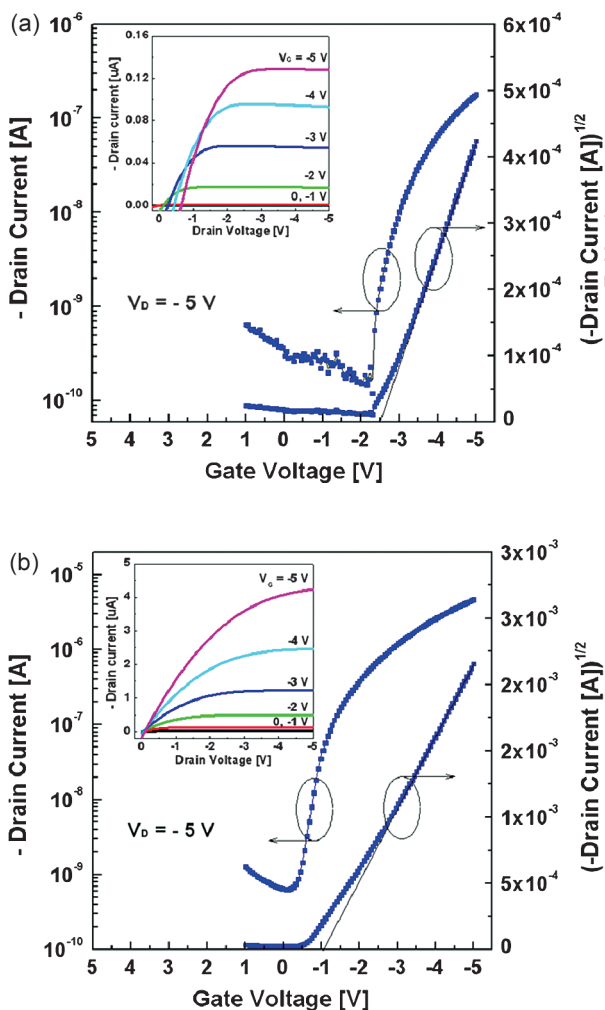
a sub-threshold slope (*SS*) of 0.07 V/dec, and a threshold voltage ( $V_{th}$ ) of  $-2.5$  V. The field effect mobility ( $\mu$ ) extracted in the saturation region was 0.02  $\text{cm}^2/\text{V}\cdot\text{s}$ , as calculated from the following equation,<sup>17</sup>

$$I_{D,SAT} = \frac{\mu_{SAT} WC_i}{2L} (V_G - V_{th})^2 \quad (1)$$

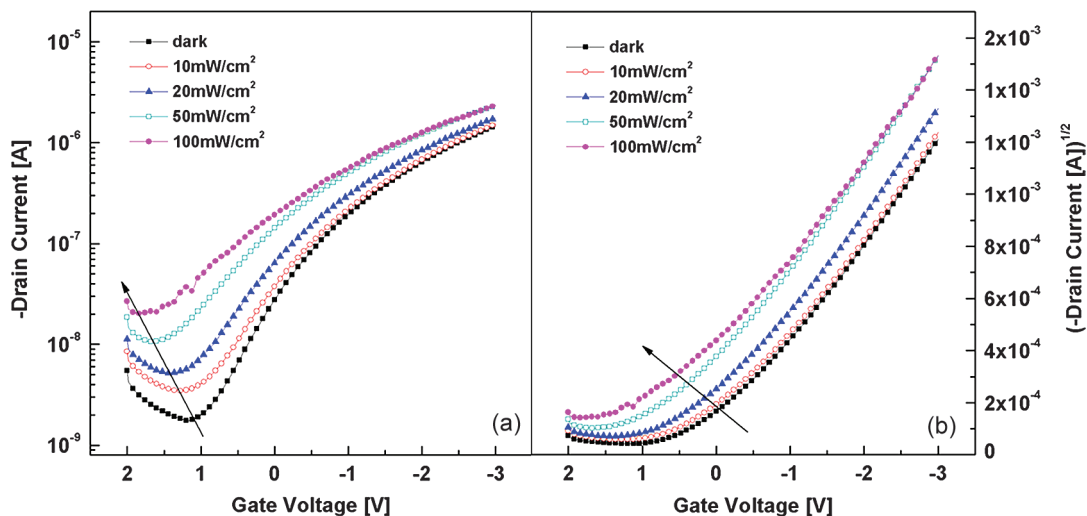
The hybrid dielectric exhibited significantly improved electrical properties, including an  $I_{on}$  current of 4.65  $\mu$ A, a current on/off ratio of  $\sim 10^4$ , an *SS* of 0.29 V/dec, and a  $V_{th}$  of  $-1.1$  V at the operating voltage of 5 V. In particular, we obtained a high  $\mu$  value of 0.72  $\text{cm}^2/\text{V}\cdot\text{s}$ , which is 36 times larger than the value for the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric, thanks to the remarkable increase in the maximum drain current. Although an excellent *SS* value has been achieved with the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric, these devices exhibit unstable characteristics and degradation in the  $I_D$  versus  $V_D$  curve. Furthermore, it should be noted that the output curve of the devices with the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric tend to shift as the gate bias increases. This is due to gate leakage and the hydrophilic nature of generally high-*k* inorganic surfaces; it is also a result of the many trap sites in the hydrophilic surface, which lead to more charge trapping as the gate bias increases, which then requires more drain voltage to achieve a negative drain current.<sup>18</sup>

As shown in the inset of Figure 2(b), this phenomenon was removed after stacking the PVP layer on the Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric, producing stable output characteristics. From the above principle, a remarkable improvement of  $V_{th}$  can be observed with such a hybrid dielectric. Figure 3(a and b) shows the transfer characteristics as the illumination was increased from 10 to 100  $\text{mW}/\text{cm}^2$ . The influence of the incident light on the devices can be clearly observed, as the overall drain currents and the threshold voltages were gradually shifted in the positive direction as the light intensity increased. This was mainly due to the creation of charge carriers during the process of photon absorption. Excitons, formed by the absorbed photons, dissociate into holes and electrons. These holes enhance the carrier density in the *p*-type OSC and reduce the threshold voltage of the device by neutralizing negatively charged states.<sup>6</sup> For this reason, the off-state drain current was significantly increased, resulting in a decrease by one order in the on/off current ratio value. It should be noted that the field-effect mobilities were nearly consistent since both threshold voltage and drain current were totally changed. In the dark, the maximum drain current at a gate bias of  $V_G = -3$  V was 1.76  $\mu$ A, compared with 2.85  $\mu$ A under illumination. In this case, the photoresponse (*R*) was 4.84 under  $V_G = 0$  V, calculated by the following equation,<sup>19</sup>

$$R = \frac{I_{D,illum}}{I_{D,dark}} \quad (2)$$



**Fig. 2.** Transfer characteristics ( $\log(-I_D)$  vs.  $V_G$  and  $\sqrt{-I_D}$  vs.  $V_G$  curves) and output characteristics ( $I_D$  vs.  $V_D$  curves, inset) of pentacene TFTs: (a) with the single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric, (b) with the hybrid dielectric.

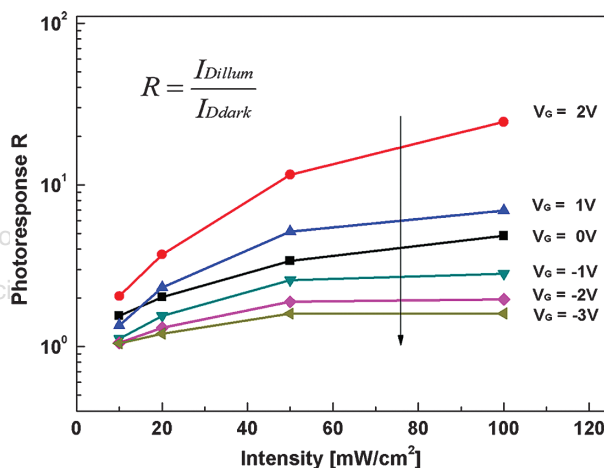


**Fig. 3.** (a)  $I_D$ - $V_G$  and (b)  $\sqrt{I_D}$ - $V_G$  transfer characteristics of the devices at different light intensities in the dark and under AM 1.5 illumination intensity from 10 to 100 mW/cm<sup>2</sup>.

In Refs. [20 and 21], off-currents of  $\sim 2.6 \times 10^{-8}$  A and  $\sim 7.0 \times 10^{-10}$  A were exhibited in the two saturation regions, respectively.<sup>20,21</sup> On the point of photo-response, we can assume that the carrier generated by photo-illumination ( $N_{\text{photo-illumination}}$ ) is constant; as such, the effect of the photo-response depending on the change of total carrier-density ( $N_{\text{total}}$ ) could be easily explained. In other words, differences of off-currents in the two devices mean that the total carrier densities of the active-layers of the two devices are different. Thereby, the effect of  $N_{\text{photo-illumination}}$  on the photoresponse is also different. The correlation of photo-carrier, bulk-carrier ( $N_{\text{bulk}}$ ) and  $N_{\text{total}}$  can be expressed as follows:

$$N_{\text{total}} = N_{\text{bulk}} + N_{\text{photo-illumination}} \quad (3)$$

In Eq. (3), since we assumed  $N_{\text{photo-illumination}}$  is constant, the effect of  $N_{\text{photo-illumination}}$  can be explained by comparing it with  $N_{\text{bulk}}$ . With the increasing  $N_{\text{bulk}}$ ,  $N_{\text{photo-illumination}}$  makes a lower contribution to  $N_{\text{total}}$ . On the other hand, with the decreasing  $N_{\text{bulk}}$ ,  $N_{\text{photo-illumination}}$  makes a greater contribution to  $N_{\text{total}}$ . Finally, the enhanced photoresponse ( $R = 4.84$ ) originate from a carrier density of active layer lower than that shown in our previous work ( $R = 1.45$ ). Photoresponse increases with increasing intensity in the off-state ( $\sim V_G < 0$  V), while the  $R$  value saturates with increasing intensity in the saturation region. In our experiments, no more photocurrent effect was observed over 100 mW/cm<sup>2</sup> of illumination intensity. It is important that most of the graphs show that the photoresponse in the off state is higher than it is in the accumulation region. This phenomenon can be explained by the fact that the photocurrent or carrier creation by the incident light is highly effective under low gate bias, and the photo-effect decreases if the behavior of the device is controlled by the gate bias in the strong-accumulation region.



**Fig. 4.** Photoresponse versus illumination intensity for various  $V_G$ .

#### 4. CONCLUSION

In summary, low-voltage-driven pentacene TFTs have been fabricated using organic and high-*k* inorganic hybrid dielectrics. We have shown that a high capacitance density can be obtained, as well as good dielectric surface properties, where the latter leads to a higher  $\mu$  than that of pentacene TFTs with a single Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> dielectric. Consequently, the cross-linked PVP/Bi<sub>5</sub>Nb<sub>3</sub>O<sub>15</sub> hybrid dielectric shows better dielectric properties, and the proposed device shows superior transistor characteristics at low voltage ( $\sim 5$  V). Furthermore, we obtained a remarkable photoresponse, including several parameter shifts under AM 1.5 illumination. These results indicate that this type of hybrid dielectric, based on an organic/high-*k* inorganic material, is a very promising candidate for use in portable electronics, having the advantage of room temperature fabrication processing in plastic electronics. Moreover, devices with hybrid dielectrics show prominent suitability for use

in optoelectronics and phototransistors, which require low power consumption.

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