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SWCNTs thin film transistor circuits fabricated on flexible substrates with inkjet-printed silver electrodes for photodetectors

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Abstract

We demonstrate thin film transistors (TFTs) and PMOS inverter circuit using drop casted single-wall carbon nanotubes (SWCNTs) as active semiconductor and spin-coated poly(4-vinylphenol) (PVP) dielectric on a flexible polyethylene naphthalate (PEN) substrate of 125- μm -thick. All the electrodes were inkjet-printed using silver ink. The fabricated TFTs exhibit a linear field-effect mobility of $2.43 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, while the saturated-load PMOS inverter achieves a voltage gain of 1.5 at a supply voltage of -15 V. The optoelectronic characteristics of these TFTs are systematically investigated under white-light illumination by monitoring drain-current (I_D) transients at different gate voltages (V_{GS}). The transfer characteristics reveal a reversal in hysteresis direction between dark and illuminated conditions, indicating that photo-induced charge-carrier recombination governs the device response. Under positive gate bias, the TFTs exhibit a pronounced and fully reversible photoresponse, with the drain current recovering completely after the light is switched off. In contrast, under negative gate bias, illumination suppresses the drain current, and the stretched-exponential transient decay becomes

progressively faster with increasing negative V_{GS} , reflecting Fermi-level-dependent carrier dynamics.

Keywords: Carbon nanotubes; Photodetectors; Thin film transistors; Flexible electronics; Inkjet printing

1. Introduction

Solution-processed SWCNT-based TFTs have attracted significant attention in flexible electronics owing to their potential applications in photodetectors, logic circuits, and other low-cost, large-area electronic systems [1-6]. SWCNTs are one-dimensional nanomaterials with excellent electrical conductivity and outstanding mechanical flexibility, enabling their dispersion in a variety of solvents for solution-based device fabrication. Among the various solution-processing techniques, spin coating method, drop casting, and inkjet printing are widely employed for the fabrication of SWCNT-based devices and circuits [7-10]. Inkjet printing is a non-contact, mask-less, and digitally controlled technique that significantly reduces fabrication costs by minimizing material wastage and enabling direct patterning over large-area substrates. In contrast, spin coating offers the advantage of producing highly uniform thin films with controlled thickness, making it particularly suitable for achieving reproducible device performance [11-14].

SWCNTs possess a direct and tunable bandgap, with the bandgap energy being inversely proportional to the nanotube diameter [15-17]. Owing to their unique optoelectronic properties, semiconducting SWCNT-based photodetectors have attracted considerable attention and are generally classified into three categories according to their photoresponse mechanisms: photoconductive, bolometric, and photodesorption-based photodetectors [18-20]. For example, S. Zhang *et al.* investigated the photoresponse of SWCNT-based TFTs under high-intensity laser

illumination ($\sim 4 \text{ W cm}^{-2}$) and demonstrated that semiconducting SWCNTs exhibit an increase in drain current upon illumination, whereas metallic SWCNTs show a decrease in drain current due to their distinct charge transport mechanisms [21]. Although SWCNT-based photodetectors have been extensively studied for ultraviolet, visible, and near-infrared light detection over a broad spectral range (400–1200 nm) on various substrates [22–24], the photoresponse mechanisms in fully solution-processed SWCNT-based photodetectors remain insufficiently understood. A comprehensive understanding of these mechanisms is therefore essential for the development of high-performance, low-cost, and flexible optoelectronic devices.

In this work, we investigate the gate-controlled photoresponse of fully solution-processed SWCNT-based TFTs fabricated on flexible PEN substrates. PMOS inverter circuits with different channel width-to-length (W/L) ratios were also realized to evaluate circuit-level performance. The V_{GS} -dependent photoresponse of these TFTs was examined under high-intensity white LED illumination, demonstrating effective modulation of the photocurrent through the applied V_{GS} . UV–visible absorption spectroscopy revealed a strong absorption peak near 400 nm, confirming the efficient light-harvesting capability of the SWCNT ink. Both steady-state and transient photoresponse measurements exhibited similar V_{GS} -dependent drain-current characteristics under illumination. The solution-processed SWCNT phototransistors demonstrated rapid response and complete recovery during repeated light on/off cycles, highlighting their excellent switching characteristics.

2. Experimental details

SWCNT-based TFTs and PMOS inverter circuits were fabricated entirely through solution-processing on flexible PEN substrates ($3.5 \text{ cm} \times 3.5 \text{ cm}$, $125 \text{ }\mu\text{m}$ thick). Prior to device fabrication, the PEN substrates were sequentially cleaned with deionized (DI) water and

isopropyl alcohol (IPA), followed by ultrasonication for 5 min. The cleaned substrates were then dried under a high-purity nitrogen atmosphere. All the electrodes were printed using a Dimatix DMP-2850 printer. The cross-linked poly(4-vinylphenol) (PVP) gate dielectric of thickness 420 nm was achieved by spin coating method. Commercial SWCNT ink dispersed in toluene and drop-cast in the channel with 0.2 μL volume and subsequently annealed at 150 $^{\circ}\text{C}$ on a hot plate to evaporate the residual toluene solvent and improve film quality. A detailed description of the device fabrication process has been reported in the previous research work [25]. Electrical characterization of the fabricated TFTs and circuits was performed using Keithley 2410 and Keithley 2636A source measurement units (SMUs). The thickness and surface roughness of the deposited thin films were measured using a Bruker DektakXT L01CL93 stylus profiler.

3. Results and discussion

The schematic device structure used in this research has been illustrated in Figure 1(a) and the I_D - V_{DS} characteristics of the fabricated TFTs are illustrated in Figure 1(b) for the maximum operating voltage of $V_{DS} = V_{GS} = -15 \text{ V}$. The corresponding transfer characteristic ($|I_D|$ - V_{GS}) in semi-log scale for $V_{DS} = -10 \text{ V}$ is depicted in Figure 1(c). High on current and negligible hysteresis was obtained which shows the good quality dielectric-semiconductor interface formation. We used Figure 1(c) for current on/off ratio and sub-threshold swing calculation. The inset in the figure shows the camera image of the flexible nature of the PEN substrate and devices fabricated on these substrates. Figure 1(d) illustrates the $\sqrt{I_D}$ vs V_{GS} curve used for the measurement of threshold voltage (V_{th}) and charge carrier mobility (μ) calculation using the equation:

$$\mu_{lin}(V_{GS}) = \frac{L}{WC_i} \frac{1}{V_{DS}} \left[\frac{\partial I_{DS}}{\partial V_{GS}} \right] \quad (|V_{DS}| < |V_{GS} - V_{th}|) \quad (1)$$

where $L=60\text{ }\mu\text{m}$ is the channel length, $W=1200\text{ }\mu\text{m}$ is the channel width, and C_i is the dielectric capacitance. We fabricated a separate Ag/PVP/Ag (MIM) sandwich structure and measured the capacitance for 420 nm-thick PVP film spin coated between inkjet-printed top and bottom Ag electrodes using a semiconductor parameter analyzer (KEYSIGHT B1500A). We achieved the capacitance value of 5.6 nF/cm^2 at 10 kHz. The sub-threshold swing (SS) determines the ability to fast swing from off to on state and defines the required V_{GS} to change drain current of one order of magnitude and mathematically calculated from the following formula using Figure 1(c):

$$SS = \frac{dV_{GS}}{d(\log I_D)} \quad (2)$$

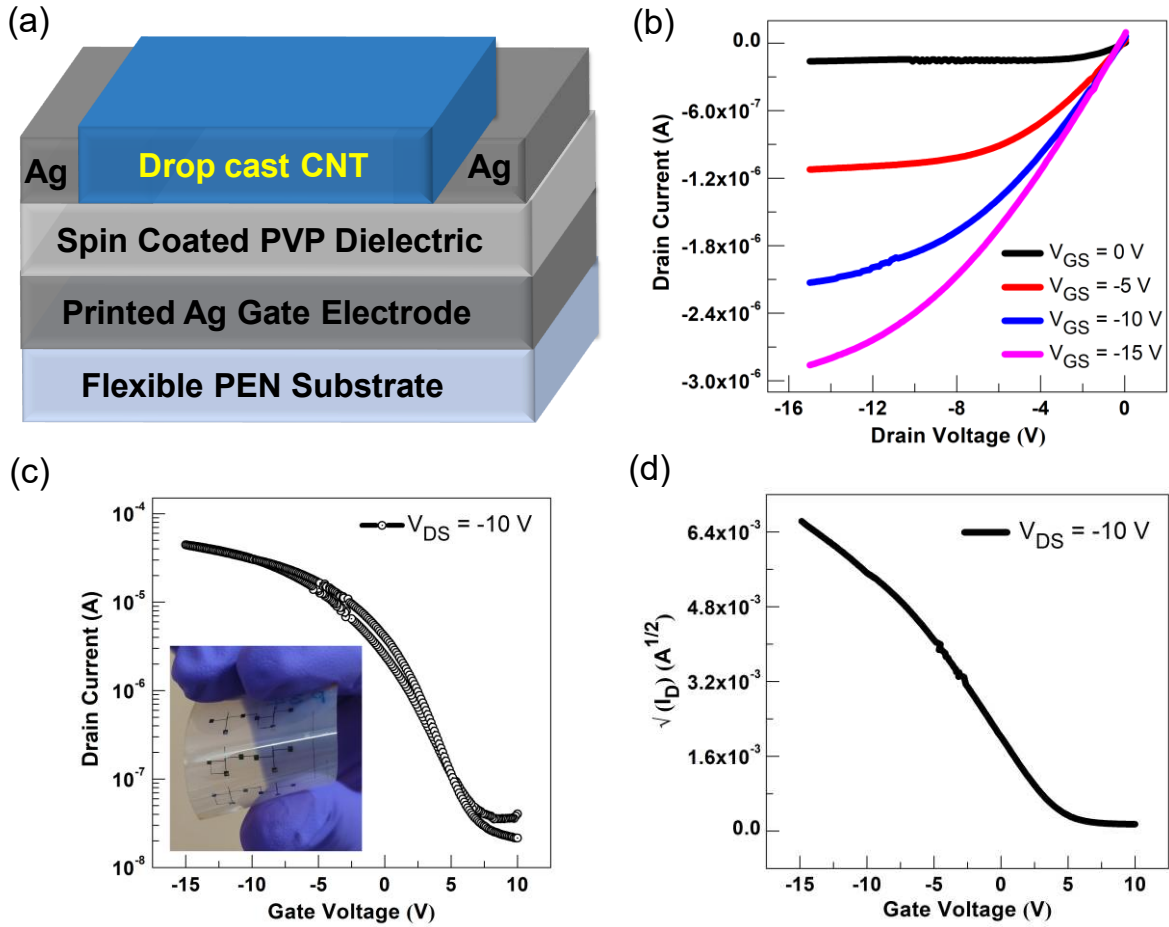


Figure 1. (a) Schematic diagram of the TFT device structure used in this research work. (b) Output characteristics for different V_{GS} steps, (c) transfer characteristics with the camera photograph in the inset, and (d) $\sqrt{I_D}$ vs V_{GS} plot at $V_{DS} = -10\text{ V}$ used for V_{Th} calculation.

The trap density (N_{it}) at the dielectric-semiconductor interface calculated using the formula:

$$N_{it} = \left[\frac{SS \log e}{k_B T / q} - 1 \right] \frac{C_i}{q} \quad (3)$$

where k_B is the Boltzmann constant and q is the charge on electron. All the TFT parameters for these CNT based devices are listed in Table 1.

Table 1. Device parameters for fully solution-processed CNT based TFT devices.

Parameter	Value
Mobility	$2.43\text{ cm}^2/\text{Vs}$
V_{th}	4.5 V
I_{ON}/I_{OFF}	2×10^3
SS	2.68 V/decade
N_{it}	$1.5 \times 10^{12}\text{ cm}^{-2}\text{eV}^{-1}$

Next to a TFT device analysis, we have fabricated a PMOS inverter circuit using inkjet printing technique. The schematic diagram for the fabricated saturated-load PMOS inverter circuit is shown in Figure 2(a) in which the channel width of the load and driver transistors are $600\text{ }\mu\text{m}$ and $1200\text{ }\mu\text{m}$ respectively, while the channel length was kept fixed $60\text{ }\mu\text{m}$ for both the TFTs. A clear camera photograph image of the fabricated PMOS flexible inverter circuit was shown in Figure 2(b). The voltage transfer characteristics (VTC) of the inverter circuit is shown

in Figure 2(c) for which the input voltage (V_{IN}) varies up to -15 V and the output voltage (V_{OUT}) was measured for different supply voltages (V_{DD}). A perfect behavior of an inverter was observed in which the V_{OUT} changes from low to high value corresponding to high and low V_{IN} voltages, respectively. The trip point ($V_{IN} = V_{OUT}$ condition) was controlled by the V_{th} and channel widths of both the transistors. The gain ($\frac{dV_{OUT}}{dV_{IN}}$) was calculated from Figure 2(c) and plotted in Figure 2(d) against the V_{IN} for different V_{DD} . We obtained the gain values of 1.2 for $V_{DD} = -15$ V.

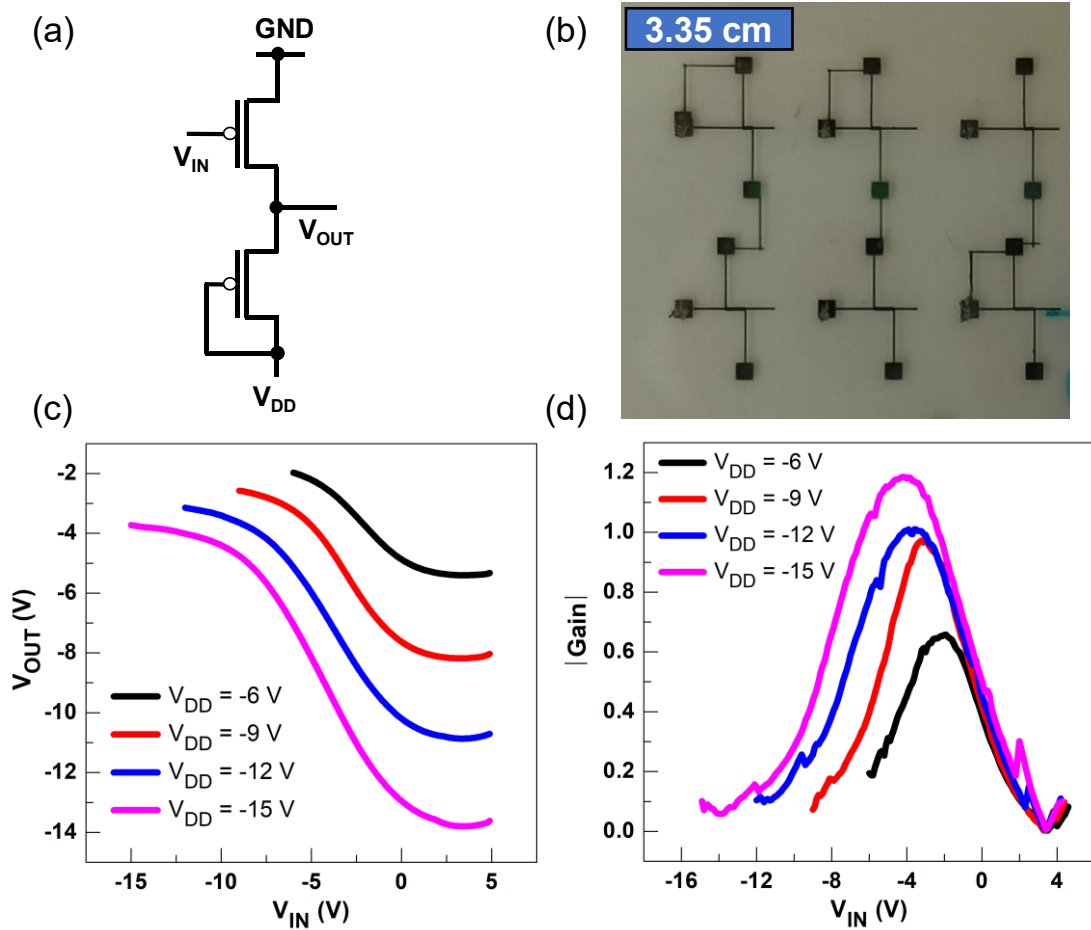


Figure 2. (a) Schematic circuit line diagram of the saturated-load inverter, (b) camera photograph of inkjet-printed PMOS inverters fabricated on PEN flexible substrate, (c) voltage

transfer characteristics, and (d) change in voltage signal gain with V_{IN} for various supply voltages.

The photoresponse behavior of these solution-processed CNT based TFT devices have been shown by using the white light LED of 3 Watt power. The distance between the light source and TFT device was kept fix 3.5 cm in all the photoresponse experiments. Figure 3(a) illustrates the $|I_D|$ - V_{GS} characteristics in dark. The increase in I_D (clockwise hysteresis) was observed and it is widely claimed that the slow polarization of dipoles presented in the bulk of the polymer dielectric enhance the net field in the channel with the applied gate voltage which in turn responsible to increase the drain current [26]. For the light experiment shown in Figure 3(b), firstly the light source was kept on for 90 seconds and then the $|I_D|$ - V_{GS} measurements were carried at $V_{DS} = -5 V$ and the light source was on in the entire dual sweep. The opposite hysteresis behavior (anti-clockwise hysteresis) was obtained with decrease in I_D . The recombination of electron-hole pair during the reverse sweep of V_{GS} is the reason for decrease in I_D . A fair comparison between dark and light $|I_D|$ - V_{GS} curves is shown in Figure 3(c). A tradeoff between gate field and light intensity was found till $V_{GS} = -7.8 V$, after which the I_D decrease in comparison to dark current due to photo generated charge carrier recombination. A similar trend in photoresponse of I_D change was verified with the transient experiments and discussed in greater details in further section.

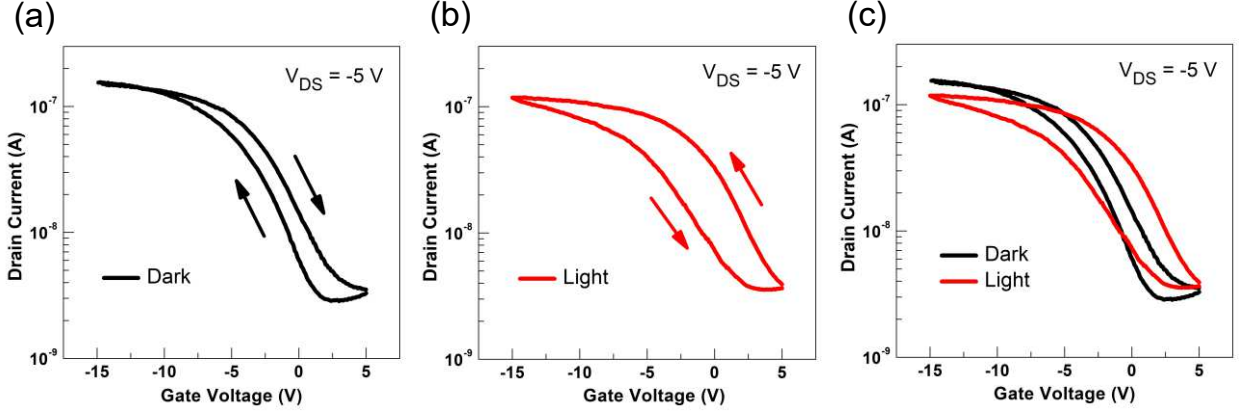


Figure 3. Transfer characteristics ($|I_D|$ - V_{GS}) (a) in dark, (b) in presence of white light LED (power = 3 Watt), and (c) combined plot of (a) and (b) at $V_{DS} = -5 V$ for flexible SWCNTs based TFTs using PVP dielectric.

In addition to steady-state characteristics we also demonstrated the dynamic photoresponse behavior of these flexible inkjet-printed SWCNTs based TFT devices. Figure 4(a) shows the UV-visible absorption spectra of the SWCNT ink with the concentration of 0.01 mg/mL in toluene. A sharp absorption peak was observed in the ultraviolet region at 399 nm and also a decreased peak in the visible region between 400 to 600 nm used for band gap calculation. The band gap value for the SWCNT was calculated using Tauc relation $(\alpha h\nu)^\gamma = A(h\nu - E_g)$, where α is the absorption coefficient, h is the Planck's constant, ν is the photon's frequency, A is the proportionality constant, and E_g is the band gap energy. Gamma (γ) denotes the nature of the electronic transition gives the value $\gamma = 2$ for direct allowed transitions, and $\gamma = 1/2$ for indirect allowed transitions. The $(\alpha h\nu)^2$ vs $h\nu$ plot is shown in the inset of Figure 4(a) and the band gap value was obtained 2.94 eV confirm the direct allowed transition in these SWCNTs. The dynamic photoresponse behavior is shown in Figure 4(b) for $V_{GS} = 0 V$. The light was switch on for 4 minutes and the increase in I_D at $V_{GS} = 0 V$ was obtained only due to the absorption of the

white light by the active CNT thin layer. Then the light source was switched off and I_D transients were measured for more than 5 minutes. After switching the light source off, the photocurrent was recovered to the initial dark current level due to the recombination of electron and holes. The fast recovery indicates the application of these CNT based TFT devices for a good photo detector or photo sensor. Figure 4(c) illustrates similar photo response characteristics in off-state for three different gate voltages i.e. $V_{GS} = 5, 7.5 \text{ and } 10 \text{ V}$. In off-state, after switching off the light source the photocurrent recovered very fast as compared to $V_{GS} = 0 \text{ V}$. We calculated the time constant for decaying transients by fitting all the dark current transients after switch off the light source using the exponential decaying function $I_D = C_1 + A_1 \exp[-(t/\tau_1)]$, where C_1 and A_1 are constants, and τ_1 is the time constant. The inset of Figure 4(c) illustrates the excellent fitting of the experimental dark current decay curve for $V_{GS} = 5 \text{ V}$ using the above exponential decaying function. We obtained the time constant values of 72, 73 and 76 s for $V_{GS} = 5, 7.5 \text{ and } 10 \text{ V}$ respectively. The lower value of time constants gives information about the fast recombination of electron-hole pairs after switch off the light source because the Fermi level move toward the valence band in comparison to $V_{GS} = 10 \text{ V}$. Since, V_{th} is positive (+4.5 V), a large number of holes at $V_{GS} = 0 \text{ V}$ are present in the semiconductor channel even after switched off the light gives slower decay transient with larger value of time constant (140 s) confirm that the recombination is a slow process for $V_{GS} = 0 \text{ V}$. The time constant values changes negligible (from 72 to 76 s) for higher positive V_{GS} values (off states) after the threshold voltage.

Figure 4(d) shows the photoresponse transients for these flexible TFT devices in the on states for $V_{GS} = -5, -7.5 \text{ and } -10 \text{ V}$ by keeping $V_{DS} = -5 \text{ V}$ fixed throughout the experiment. In the dark state, the increases in I_D is due to slow polarization of the dipoles present in the polymer dielectric by applying the negative gate voltages. The drain current decreases when the

light source was switched on. In the on state, a large number of holes are available in the semiconductor channel by applying negative V_{GS} due to which Fermi level moves closer to the valence band and give a fast recombination. We fitted these decaying transients with stretched exponential function $I_D = C_2 + A_2 \exp\left[-\left(\frac{t}{\tau_2}\right)^\beta\right]$, where, C_2 and A_2 are constants, τ_2 is the time constant, and β is the stretched exponential factor. We obtain all the parameters from the curve fitting for $V_{GS} = -5, -7.5$ and -10 V, and listed in the Table 2.

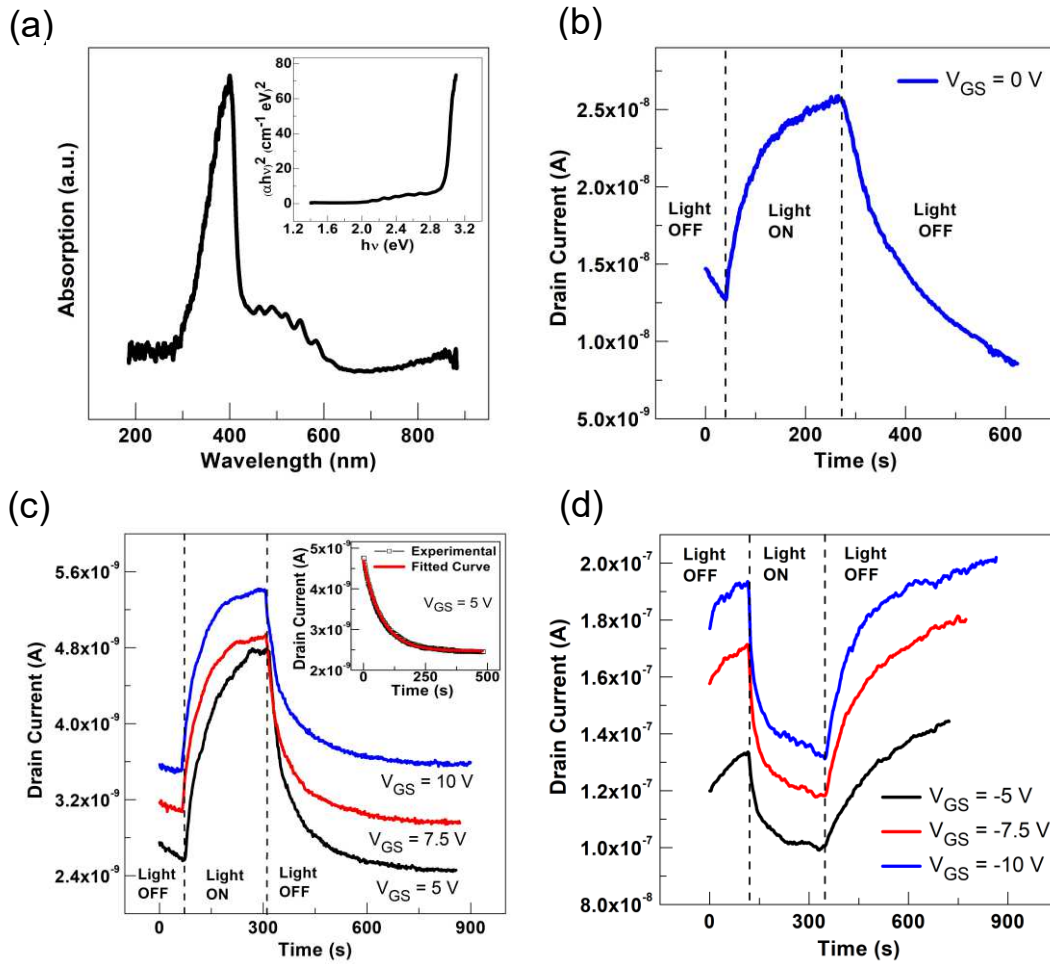


Figure 4. (a) UV-visible absorption spectra of the SWCNT ink (0.01mg/mL) in toluene. Dark and photo-current measurements as a function of time at fix $V_{DS} = -5$ V for (b) $V_{GS} = 0$ V, (c)

three different positive (off state) gate voltages i. e. $V_{GS} = 5, 7.5, \text{ and } 10 \text{ V}$, and (d) three different negative (on state) gate voltages i. e. $V_{GS} = -5, -7.5, \text{ and } -10 \text{ V}$.

Table 2. Fitting parameters for $V_{GS} = -5, -7.5 \text{ and } -10 \text{ V}$ using stretched exponential curve.

$V_{GS} \text{ (V)}$	$\tau_2 \text{ (s)}$	β	A_2	C_2
-5	32.4	0.62	3.79×10^{-8}	9.92×10^{-8}
-7.5	31.2	0.56	6.06×10^{-8}	1.16×10^{-7}
-10	20.3	0.44	7.85×10^{-8}	1.31×10^{-7}

From Table 2 it is clear that the time constant (τ_2) reduced from 32 s to 20 s when V_{GS} changes from -5 to -10 V. The Fermi level is much close to the valence band for $V_{GS} = -10 \text{ V}$ which gives the fast recombination with the low time constant of 20 s. This clearly indicates that the photoresponse become faster for larger negative V_{GS} value. Also the transients becomes more non exponential with lower values of β .

4. Conclusions

In summary, we have fabricated high mobility SWCNTs based TFT devices and saturated-load PMOS inverter circuits on 125 μm -thick flexible PEN substrate using fully solution-processed techniques. We achieve the mobility of $2.43 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and PMOS inverter gain of 1.5 for driver-to-load channel width ratio of 2. The application of these solution-processed TFT devices as a white light photodetector has been shown for positive and negative V_{GS} . Both the steady-state and dynamic characteristics of TFT devices shows that during white light illumination, the I_D increases in the off state due to optical absorption and decreases in the on state because of the recombination of charge carriers. After switch of the light source, the

photocurrent transients become slower for larger positive V_{GS} and faster for more negative V_{GS} values while the light source is switched on due to movement of Fermi level close to valence band.

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Data Availability Statement

All data generated or analysed during this study are included in this published article.

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