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Graphene-lead zirconate titanate optothermal field effect transistors

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We have developed a pyroelectric field effect transistor (FET) based on a graphene-lead zirconate titanate (PZT) system. Under the incidence of a laser beam, the drain current can be increased or decreased depending on the direction of the polarization of the PZT substrate. The drain current sensitivity of the optothermal FET can reach up to 360 nA/mW at a drain field of 6.7 kV/m more than 5 orders of magnitude higher than that of the photogating transistors based on carbon nanotube on SiO_2/Si substrate. Graphene is an excellent component for pyroelectric FET due to its high optical transparency and conductance. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3693607]

Recently, ferroelectric materials with up and down remnant polarizations have been employed in non-volatile ferroelectric field effect memories as gate dielectrics.[1–5] By replacing the typical gate dielectric SiO_2 with a ferroelectric material with high dielectric constant such as lead zirconate titanate (PZT), the transconductance of a field–effect transistor (FET) can be increased. For example, Hong *et al.* reported a tenfold increase of carrier mobility in few-layer graphene (FLG) field-effect transistors when the SiO_2 substrate is replaced by single-crystalline epitaxial $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT).[5] Furthermore, recent studies have shown that it is also possible to generate a gate voltage in a FET by non-electrical means, which is advantageous from an energy conservation stand point.[6,7] For example, Marcus *et al.* demonstrated the so-called photogating in which carbon nanotube transistors were gated by light through the photovoltaic voltage generated in the silicon substrate.[6] Liu *et al.* investigated piezoelectric gated hybrid field-effect transistor that consists of carbon nanotubes at the bottom and crossed ZnO fine wires on the top with an insulating layer in between.[7] It is known that PZT is a pyroelectric material whose polarization can change when temperature is varied.[8] As a result, the bound charge density on the PZT surface can change with temperature, thus generating a temperature-change induced current in an external circuit.[9–11] Combining the pyroelectricity and the high dielectric constant of PZT, it is possible to create a high-performance FET that can be gated with non-electric means.

In this communication, we report a graphene-PZT FET whose drain current can be modulated by an optothermal gating mechanism where PZT served as the gate dielectric and graphene as the drain conductor. The optothermal feature of the device is especially suitable for remote or wireless applications. The ultra-thin graphene layer was deposited by chemical vapor deposition (CVD) and was subsequently transferred to the PZT substrate.[12] An infrared (IR) laser of 1064 nm wavelength and 320 mW power was used as a light source. It was shown that the drain current can be modulated by the incidence of the IR light via the pyroelectric effect of the PZT. Furthermore, the drain current could be increased or decreased by the incident IR light by properly orienting the polarization of the PZT layer.

Figure 1(a) shows the schematic of the graphene-PZT transistor. The graphene layers used in this study were grown on a copper foil in vacuum by the CVD method in a tube furnace using methane and hydrogen gases.[12] Graphene samples were then transferred to the PZT (T105-A4E-602, Piezo Systems, Boston, MA) substrate or the SiO_2 wafer through polymethyl methacrylate (PMMA) coating and iron (III) nitrate etching.[12–14] In this experiment, the thickness of the PZT layer was 127 μm and the polarization had been poled to orient in the direction perpendicular to the surface. The as-grown graphene was transferred to the side of the PZT where the original Ni electrode was first removed by HNO_3 etching. Finally, a mask was used to define the source-drain contact pattern with a 0.15 cm channel length and 0.35 cm channel width of graphene. The Ti/Au (10/100 nm) deposition was then carried out to create the source and drain electrodes of the graphene-PZT or graphene-SiO_2 transistors. The graphene-PZT FETs were then placed on an ITO (Indium Tin Oxide) glass as a back electrode with conductive glue. The graphene-SiO_2 layers were characterized by Raman spectroscopy, and the charge mobility of graphene is estimated ~1200 $cm^2/(Vs)$ from the I_d-V_g characteristics (not shown here) and capacitance of the gate oxide.

Figure 1(b) shows the Raman spectroscopy results obtained on one of the transferred graphene samples on SiO_2. For this sample, the spectrum contained a large signal of G and 2D peaks indicating the presence of few-layer graphene.[15] We have fabricated two kinds of graphene-PZT FET: One with the polarization of the PZT pointing down (away from the graphene layer) and the other with the polarization of the PZT pointing up (towards the graphene layer).

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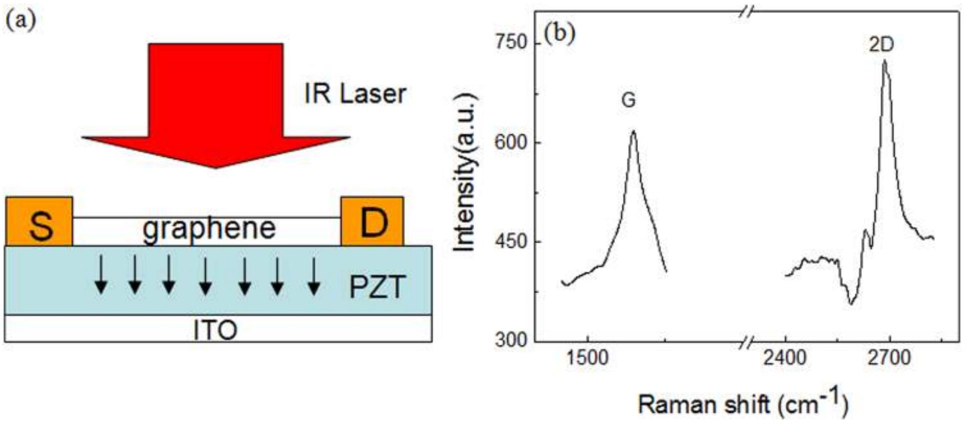

FIG. 1. (Color online) (a) The schematic diagram of the graphene-PZT field effect transistor. (b) Raman spectra of the transferred graphene on SiO_2.

The drain current versus gate voltage (I_d vs. V_g) at a constant drain voltage (V_d) of a graphene-PZT FET with a down polarization and that of a graphene-PZT FET with an up polarization as measured with a Keithley instrument are shown (black filled squares) in Figs. 2(a) and 2(b), respectively. Because the properties of graphene layers varied, the V_d for Fig. 2(a) is 0.7 V while that for Fig. 2(b) is 0.015 V. As can be seen in both Figs. 2(a) and 2(b), the drain current decreased with an increasing gate voltage from −25 V to 25 V indicating that the graphene is a doped p-type semiconductor which is common in as-fabricated graphene devices.[16] We further investigated the graphene-PZT transistors in the presence of the IR laser beam. The resultant I_d vs. V_g with IR illumination is also plotted in Figs. 2(a) and 2(b) (red filled circles). As can be seen, the drain current of the FET with a down polarization shifted down under the IR illumination of 100 mW while that of the FET with an up polarization shifted up under IR illumination.

To understand the behaviors in Fig. 2, we note that the polarization of a piezoelectrics such as PZT changes with temperature. An increase in temperature causes the spontaneous polarization P_S to decrease as the average magnitude of dipole moment diminishes.[17–19] Furthermore, there were bound charges on the surfaces of the PZT. A top surface of a PZT with a down (up) polarization had bound negative (positive) charges. When the p-type graphene was placed on top of the PZT substrate with a down polarization, the graphene was in contact with the bound negative charges on the PZT surface, which would help concentrate the hole carriers in the graphene layer. When illuminated by the IR laser beam,

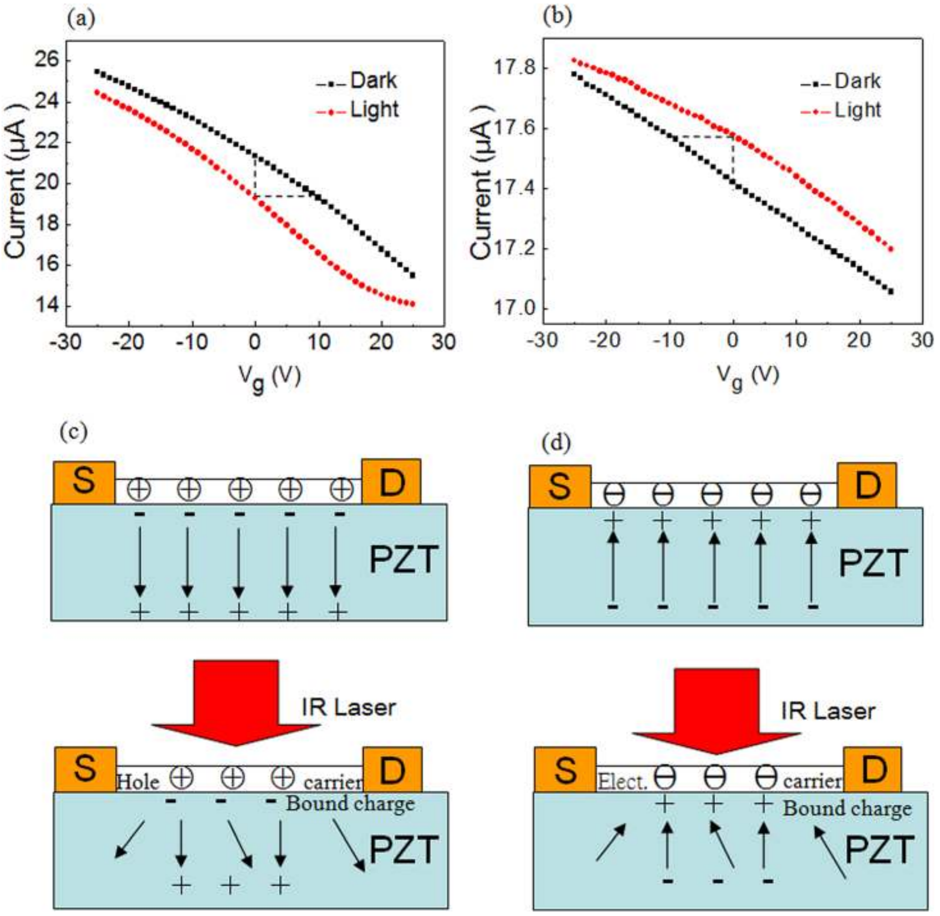

FIG. 2. (Color online) The drain current as a function of gate voltage corresponding to dark (black filled square) and light (red filled circle) states under 100 mW IR illumination for the device with (a) downward and (b) upward polarizations. (c) IR illumination on a PZT with a down polarization would decrease the negative bound charge density at the top surface of the PZT (due to the decreased polarization). As a result, the hole carrier concentration in the graphene layer would decrease resulting in a decreased drain current. (d) IR illumination on a PZT with an up polarization would decrease the positive bound charge density at the top surface of the PZT, which would act to decrease the electron carrier concentration and increase the hole concentration in the graphene layer, thereby increasing the drain current.

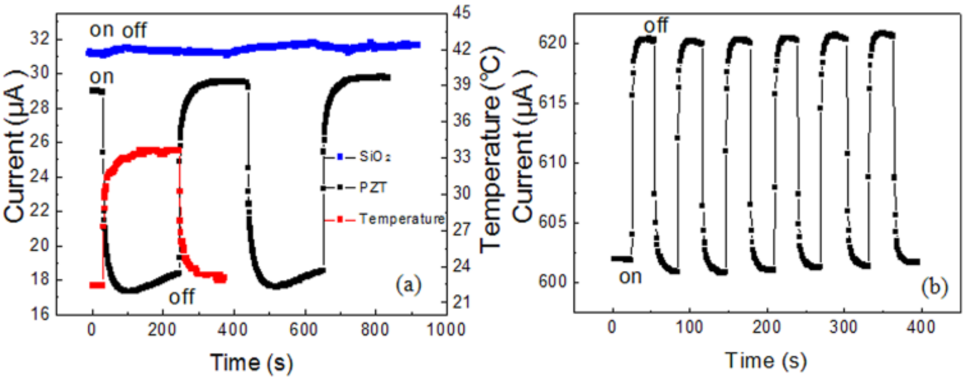

FIG. 3. (Color online) The behavior of current and temperature as a function of time modulated by IR laser of 320 mW corresponding to (a) downward and (b) upward polarizations of the PZT substrate. The frequency of the oscillation depends on the frequency of the light blocker. (a) also shows the kinetic behavior of graphene-SiO_2 system. There is very little response when IR laser was introduced in graphene-SiO_2.

the resulting temperature increase would decrease the polarization and the density of the negative bound charges at the top surface of the PZT.[9,19–22] As a result, the hole carrier concentration in the graphene layer would decrease resulting in a decreased drain current. On the other hand, IR illumination on a PZT with an up polarization would decrease the polarization and the positive bound charge density on PZT. As a result, the electron carrier concentration in the graphene layer would decrease, thus effectively increasing the hole concentration and the drain current. The behavior is illustrated as schematics in Figs. 2(c) and 2(d), respectively.

The IR illumination was also modulated with a manual light blocker. To best see the optical-modulation effect on the drain current, we plot the drain current versus time for the FET with a down polarization and that for an up polarization by inserting light blockers periodically in Figs. 3(a) and 3(b). As can be seen, the drain current of the FET with a down (up) polarization decreased (increased) when the light blocker was removed and increased (decreased) when the blocker was inserted, consistent with modulation of the I_d versus V_g shown in Figs. 2(a) and 2(b). Note that the insertion and removal of blockers was done in different time intervals in Figs. 3(a) and 3(b). Similar study was performed for graphene-SiO_2 device. The results were included in Fig. 3(a), and there was no noticeable change in the drain current under IR illumination when blocker was removed or inserted. Furthermore, the behavior of temperature change is directly correlated with the change in drain current as shown in Fig. 3(a), indicating the devices are sensitive to temperature change. Although it is possible that high temperature can affect the property of graphene directly,[23] our results show an increased and decreased drain current by laser depending on the polarizations of PZT. This clearly demonstrates that the behavior seen in Figs. 2 and 3 is due to the presence of PZT whose polarization plays a key role in the observed behavior.

The magnitude of equivalent gate voltage induced by the IR illumination can be estimated using the gate voltage

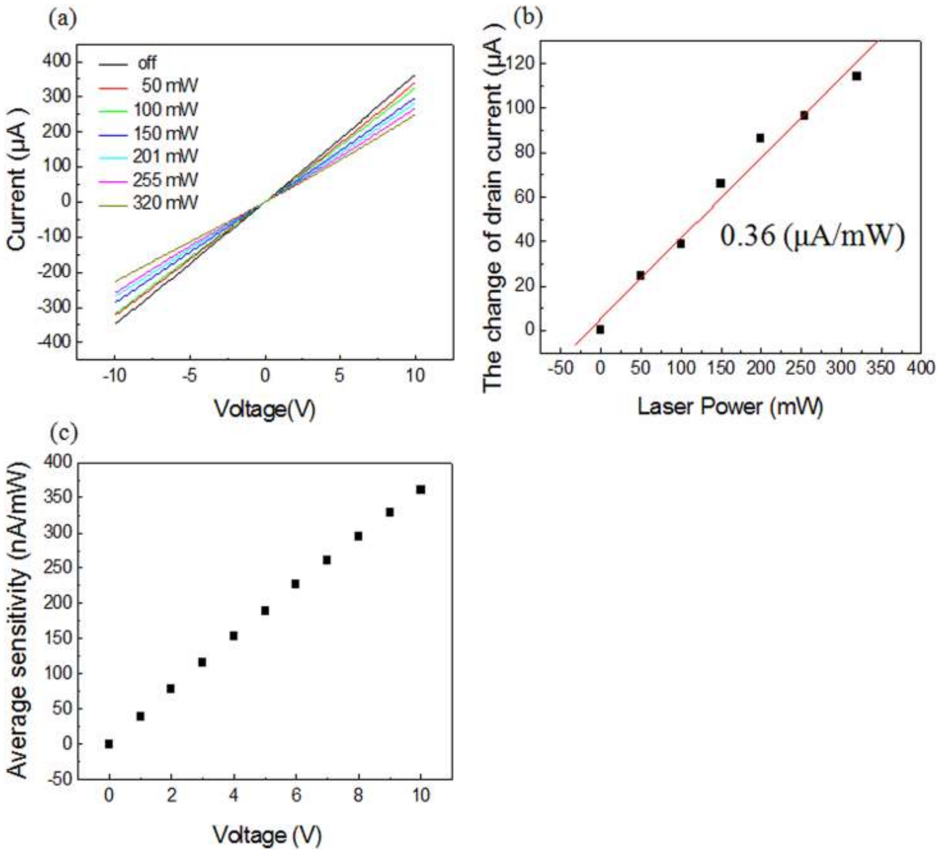

FIG. 4. (Color online) (a) The drain current as a function of drain voltage for graphene-PZT transistor with downward polarization subjected to laser of different powers. (b) The change of drain current vs laser power at an external drain voltage of 10 V. It shows that the average sensitivity is about 360 nA/mW. (c) The average sensitivity as a function of drain voltage.

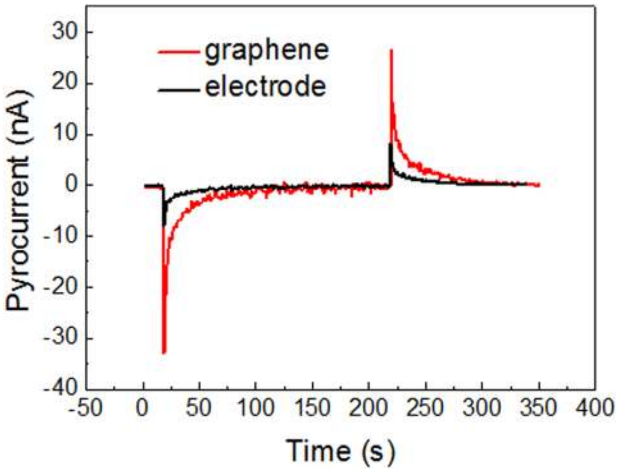

FIG. 5. (Color online) The pyroelectric current as a function of time when the laser (200 mW) was directed at the electrode and graphene, respectively.

difference between the I_d versus V_g at the same drain current with and without IR illumination. From Figs. 2(a) and 2(b), we can see that the illumination of IR at 100 mW on a PZT with a down polarization was equivalent to a gate voltage of 10 V, whereas the same IR illumination on a PZT with an up polarization was equivalent to a gate voltage of -10 V. It should be noted that the different magnitudes in drain current change between the down polarized and up polarized devices is because the graphene layers have different properties because of the difficulty in controlling the growth of large-area graphene and maintaining the same quality by the CVD method.

Figure 4(a) shows the I-V curves of graphene-PZT transistor with a down polarization illuminated by the IR laser at various powers in the range of $-10\ \text{V} < V_d < 10\ \text{V}$, where V_d is the drain voltage between the source and the drain while no V_g is applied. For pyroelectric detectors with metal electrodes, we can define the drain current sensitivity as $R_d = I_d/W$, where W is the radiation power that causes the drain current I_d to change. For our devices, the maximum current sensitivity was ~360 nA/mW at $E_d = 6.7$ kV/m, where E_d is the electric field across the drain and source electrodes. The average current sensitivity vs. drain voltage was shown in Figs. 4(b) and 4(c) which is 5 orders of magnitude higher than the typical 20 nA/W at $E_d = 50$ kV/m of the photogating transistors based on carbon nanotube on SiO_2/Si substrate.[6] Fig. 4(b) shows that the increase in drain current behaves linearly with increasing laser power indicating the optothermal gating effect is linear in nature.

Figure 5 shows the pyroelectric current (i. e., current from drain to ITO) of the graphene-PZT FET when the laser (200 mW) was incident on the drain or source electrode and graphene, respectively. In this measurement, we used source and bottom (ITO) contact of graphene-PZT transistor as electrodes similar to the traditional pyroelectric detector. It was found that the induced current of graphene is larger than that of the metal contact by about four times, indicating that the high optical transparency of graphene can enhance the device performance.

In conclusion, we have developed a graphene-PZT optothermal field effect transistor that can be tuned by laser. By applying drain voltage and laser heating, the drain current can be increased or decreased depending on the direction of polarization in PZT substrate. The optothermal gating is quite sensitive due to the high optical transparency and conductance of graphene.

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