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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 SiO2/Si substrate. Graphene is an excellent component for pyroelectric FET due to its high optical transparency and conductance.

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 SiO2 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 SiO2 substrate is replaced by single-crystalline epitaxial Pb(Zr0.2Ti0.8)O3 (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 standpoint.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. 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 SiO2 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 HNO3 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-SiO2 transistors. The graphene-PZT FETs were then placed on an ITO (Indium Tin Oxide) glass as a back electrode with conductive glue. The graphene-SiO2 layers were characterized by Raman spectroscopy, and the charge mobility of graphene is estimated ~1200 cm2/(Vs) from the Iq-Vg 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 SiO2. For this sample, the spectrum contained a large signal of G and 2D peaks indicating the presence of few-layer graphene.15

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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,
the resulting temperature increase would decrease the polar-
ization 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 illumina-
tion 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 illus-
trated 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 polariza-
tion 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 inserted, consistent with modulation of the Id
versus Vg shown in Figs. 2(a) and 2(b). Note that the inser-
tion and removal of blockers was done in different time
intervals in Figs. 3(a) and 3(b). Similar study was performed
for graphene-SiO2 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 tempera-
ture 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 demon-
strates 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

![Graph](image-url)
Figure 5. (Color online) The pyroelectric current as a function of time when the laser (200 mW) was directed at the electrode and graphene, respectively.

The 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 $-10 \text{ V}$. 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 $\sim 360 \text{ nA/mW}$ at $E_d = 6.7 \text{ kV/m}$, 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 $E_d = 50 \text{ kV/m}$ of the photogating transistors based on carbon nanotube on SiO$_2$/Si substrate.

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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