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Supplementary information

Wafer-scale graphene and ferroelectric multilayer for flexible and fast-switched modulation applications

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Electro-Optic measurement:

The method is based on the observation of frequency doubling in the detection signal when a laser beam is phase modulated with a fundamental frequency in the system. The birefringence phase shift ($\Delta\Phi$) is given by:

$$\frac{I}{I_0} = \frac{1}{4}\sin^2\left(\frac{\Delta\Phi\pi}{\lambda}\right) \quad (1)$$

where I_0 and I are the incident laser intensity and the laser intensity through EO thin films in applied electric field, respectively. The linear (Pockels effect) or quadratic (Kerr effect) EO effect alters the refractive index of the EO materials, which give rise to a shift in phase and thus convert into intensity modulation. The relationship between the phase shift ($\Delta\Phi$) and the birefringence shift (Δn) is given by:

$$\Delta\Phi = \frac{2\pi}{\lambda} l \Delta n \quad (2)$$

where λ and l are the wavelength of laser and the light path, respectively.

For these electro-optic materials, the linear and quadratic EO effects are defined as:

$$\Delta n = -\frac{1}{2}n^3 r_c E \quad \text{or} \quad \Delta n = -\frac{1}{2}n^3 R E^2 \quad (3)$$

where r_c, R, n and E are the linear EO coefficient (Pockels coefficient), quadratic EO coefficient (Kerr coefficient), refractive index, and electrical field, respectively. Based on Equation (1), (2) and (3) above, the linear EO coefficient (r_c) or quadratic EO coefficient (R) can be calculated:

$$r_c = \lambda\Delta\Phi / (\pi n^3 E L) \quad \text{or} \quad R = \lambda\Delta\Phi / (\pi n^3 E^2 L) \quad (4)$$

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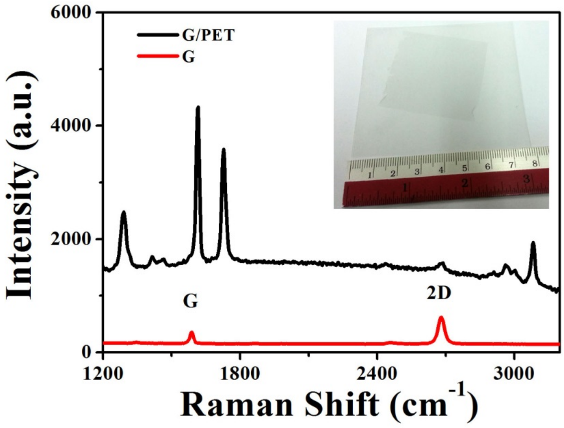

Fig. S1 Raman spectra of bare graphene and graphene/PET film. The inset is optical images of graphene film on PET substrate.

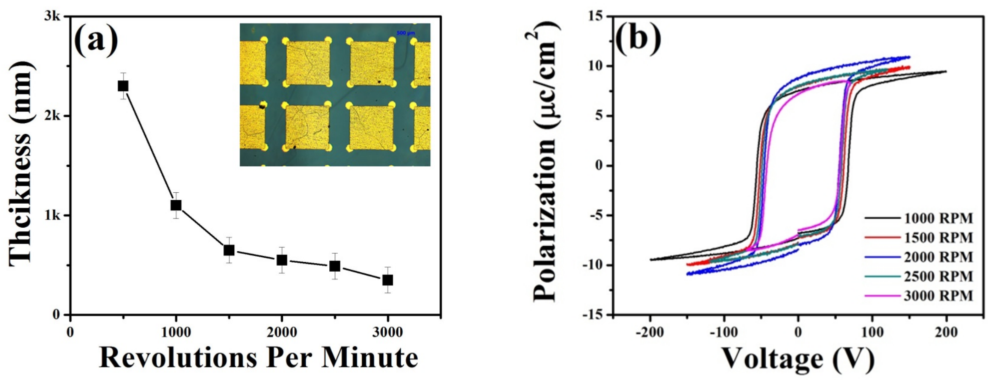

Fig. S2 (a) The thickness and (b) hysteresis polarization loops of P(VDF-TrFE) thin film versus different spin speed. The inset of (a) is G/P(VDF-TrFE) on SiO2 wafer with Ti/Au (5/100 nm) electrodes for ferroelectric measurement. In each unit cell, the graphene area is 1.44 mm^2.

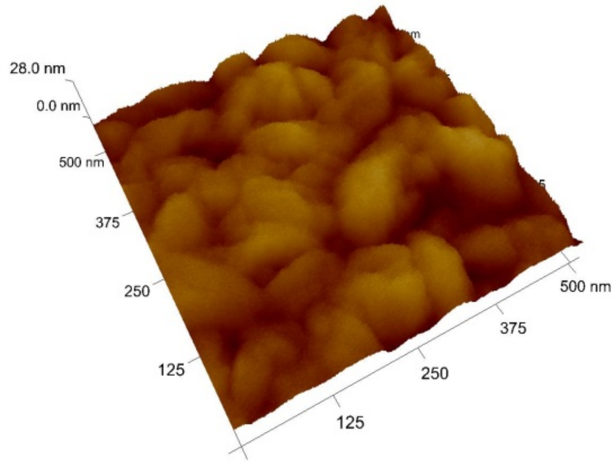

Fig. S3 Surface morphology of P(VDF-TrFE) thin film.

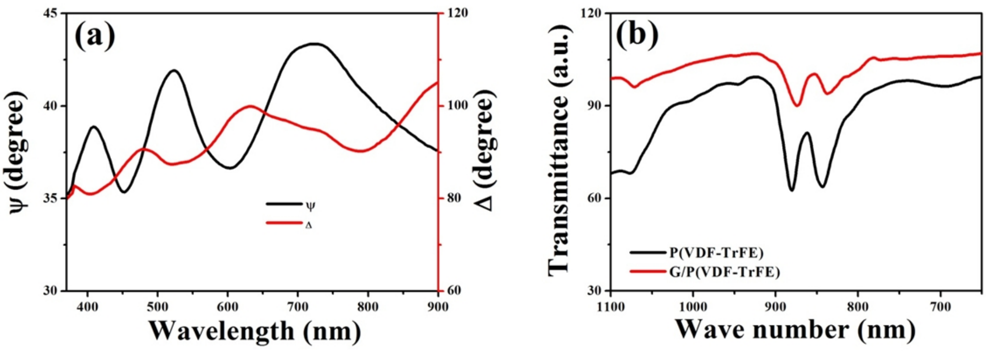

Fig. S4 (a) Spectra of the ellipsometric parameter *Ψ* and Δ as a function of wave number for P(VDF-TrFE) thin film. (b) FTIR spectra of bare P(VDF-TrFE) and G/P(VDF-TrFE) film.

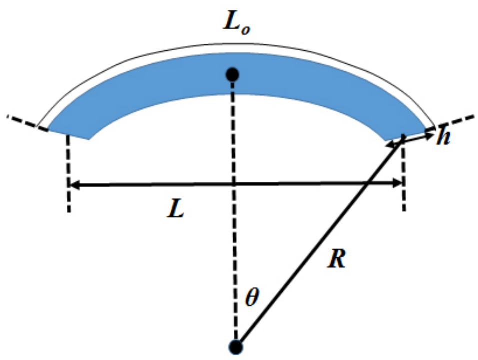

Fig. S5 Illustration of device bending under the strain.

As shown in Figure S5, the device is bent into an arc shape with a dimension. In this case, the strain in the length direction (ε) of the device is given by:

$$R.2\theta = L_o$$

$$Sin\ \theta = Sin\ L_o/2R = L/2R$$

$$\varepsilon = h/2R$$

where ***h*** is the thickness of the device (550 nm), R is the bending radius that varies between ~ 7.0 and ~ 2.0 cm. In this case, the strain (*ε*) is 0.03 -0.394%.

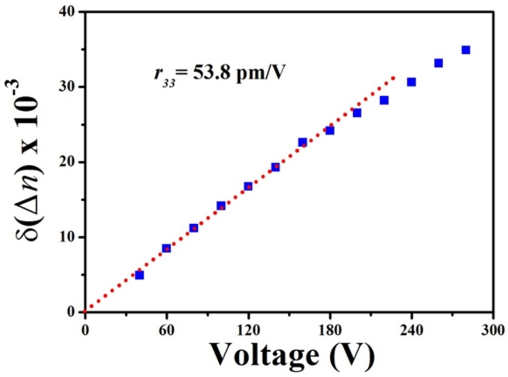

Fig. S6 Birefringence δ(Δn) in G/P(VDF-TrFE)/G multilayer film as a function of applied voltage. The thickness of P(VDF-TrFE) thin film is about 1.2 μm.

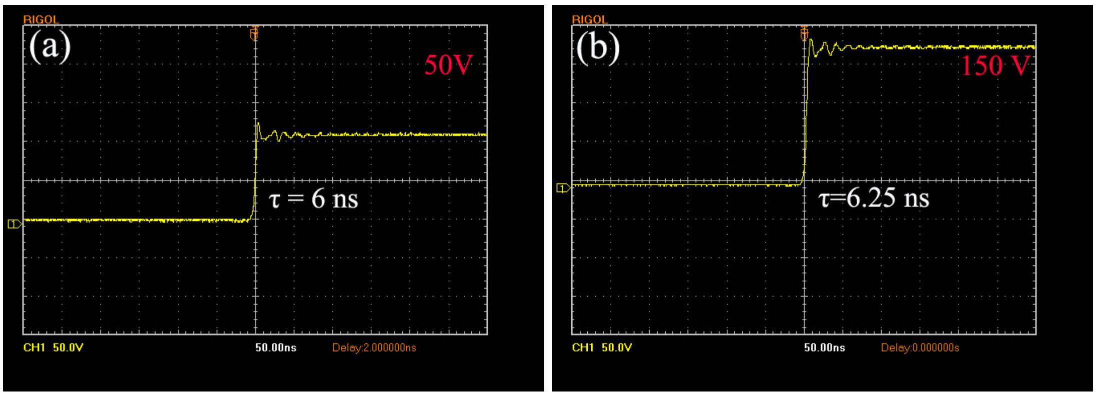

Fig. S7 The responsivity of switching time (τ) on pulse generator and power amplifier at (a) 50 V and (b) 150 V. The responsivities of switching time are 6 and 6.25 ns, respectively.