Ultrathin Multi-functional Graphene-PVDF Layers for Multi-dimensional Touch Interactivity for Flexible Displays

*Shuo Gao, Xingyi Wu, Hanbin Ma, John Robertson and Arokia Nathan**

Department of Engineering, University of Cambridge, Cambridge, CB3 0FA, United Kingdom

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ABSTRACT: This paper presents a flexible graphene/polyvinylidene difluoride (PVDF)/graphene sandwich for three-dimensional touch interactivity. Here, x-y plane touch is sensed using graphene capacitive elements, while force sensing in the z-direction is by a piezoelectric PVDF/graphene sandwich. By employing different frequency bands for the capacitive- and force-induced electrical signals, the two stimuli are detected simultaneously, achieving three-dimensional touch sensing. Static force sensing and elimination of propagated stress are achieved by augmenting the transient piezo output with the capacitive touch, thus overcoming the intrinsic inability of the piezoelectric material in detecting non-transient force signals and avoiding force touch mis-registration by propagated stress.

Traditional touch screen panels (TSPs) employ resistive, capacitive, optical and acoustic wave related architectures[1-5], to detect 2D (x-y) touch events by sensing a certain type of physical signals. For example, finger touch induced capacitance change is picked up by a capacitive TSP to recognize the presence of a touch event. Besides the limited sensing capability, Indium tin oxide (ITO) is widely used in electrodes in TSPs, in which its brittleness limits the flexibility of the touch panel[6]. Therefore, n-dimensional touch detection and high flexibility are desired attributes for future TSPs, to bring user-experience to new and advanced levels[5]. Achieving this by conventional techniques requires additional sensors and complex circuitry, resulting in extra costs and higher power consumption. Thus, it is expected that multi-functional, simple-structured devices will be used in future designs to reduce circuit complexity and power consumption, while providing multiple functions to customers.

Recent progress on piezoelectric materials provides a promising solution to achieve multi-dimensional sensing. As a non-centrosymmetric structured material, polarization is induced when subject to stress, resulting in charge generation on electrodes[7-10]. The concentration of induced charge is linear with the strength of the applied force. This enables sensing in multi-dimensions (x-y-z) [8], with high sensitivity in the z-direction because of the relatively high piezoelectric coefficient (d_{33} = 33 pC/N)[10]. In terms of flexible electrodes, materials such as metal nanowires[11-12]/nanogrids[13], Poly(3,4ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)[14], carbon nanotube (CNT)[15] and graphene have been reported. In this work, we address the use of mono-layered graphene[6,16-29], as it potentially offers possibilities of low manufacturing cost[17] due to the roll-to-roll production method[17]. Graphene also possesses strong mechanical strength (Young's modulus of 1 TPa and intrinsic strength of 130 GPa)[27]. The fracture strain of graphene can be one order higher than that of the ITO[6]. Furthermore, graphene provides the best optical transparency (97.7%)[19] among others, indicating it is a strong candidate to function as electrodes for display related applications.

In previous work[4,26,28-29], piezoelectric materials and graphene have been employed for force sensing in touch panels. However, previous studies focused on the use of the piezoelectric property of the piezoelectric materials without considering the other properties such as the electric conductivity. Furthermore, there are two critical issues associated with the nature of the piezoelectric materials in force sensing in touch panels that haven't been addressed till now. First, only detections of dynamic force signals have been reported. Second, fake force touch signals can be generated due to stress propagation from the actual force touch location(s). Thus it is necessary to distinguish the fake from real signals. This has been one of the big limitations in successful use of piezoelectric based touch interactivity.

In this article, we demonstrate a touch screen system using a graphene and polyvinylidene difluoride (PVDF) based ultra-thin film (~40 μm) for concurrently sensing two different external physical stimuli, by employing both the piezoelectric and the electrical conductive properties of the materials in the stack-up, which paves a new pathway to detect multiple stimuli. The film is structured in a graphene/PVDF/graphene sandwiched architecture. Large area monolayer graphene is used as a flexible and transparent electrode. It is prepared using the previously reported chemical vapor deposition (CVD) recipe[18]. Remarkably, the average graphene domain size reaches as large as ~1 mm[18]. The domain boundary is thereby suppressed so that the conductivity is homogeneously high over practical device scales[18]. Commercial PVDF is employed as an insulating layer for the graphene capacitor, and as a force sensing layer due to its piezoelectric property[7] to detect applied force. Hence, this extremely simple device architecture can simultaneously achieve both capacitive and force sensing, as conceptually depicted in Fig. 1

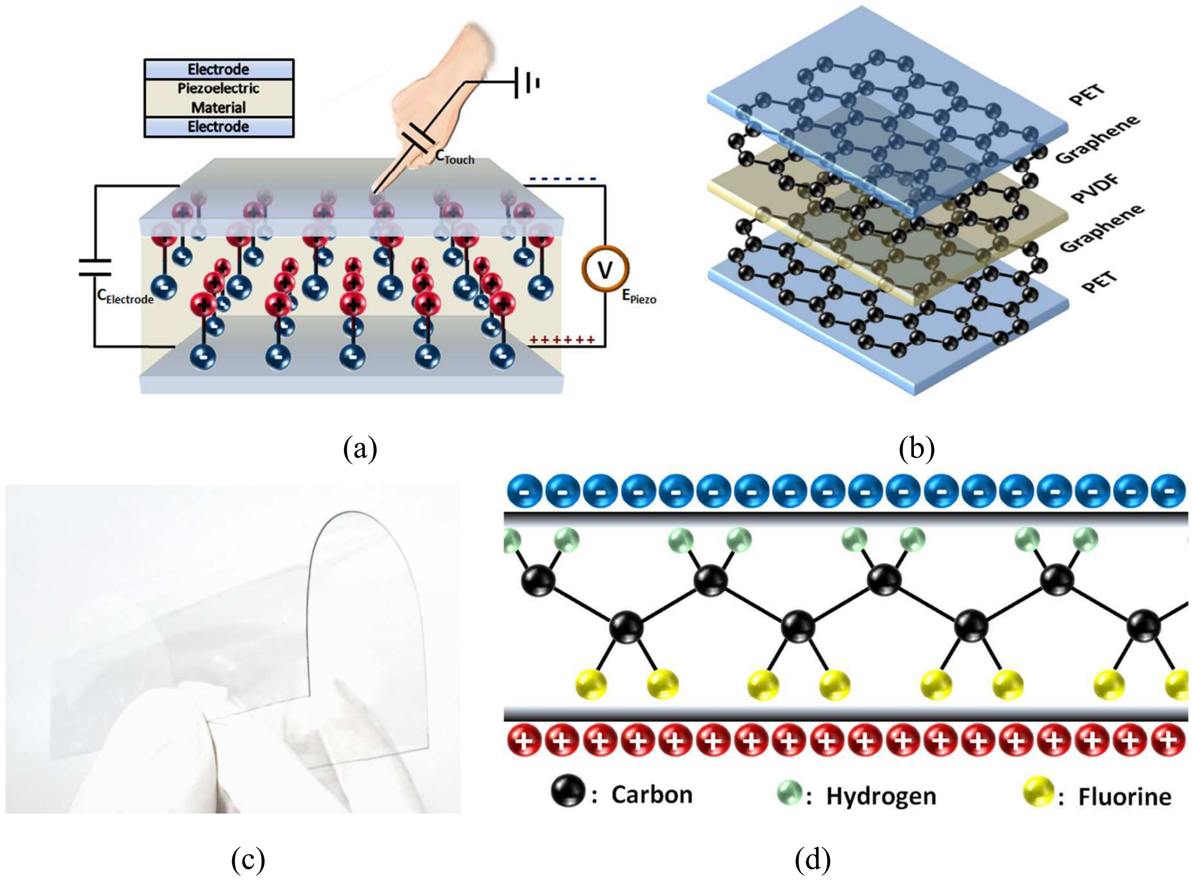

Figure 1. Stack-up of the proposed (a) multi-functional film and (b) touch panel. (c) Photograph of the graphene/PVDF ultra-thin multi-functional touch panel. (d) Structure representation and working principle of β-phase PVDF.

(a). Experimental results have shown that the device provides high sensitivity in both capacitive and force touch detection modalities, together with good mechanical durability.

A touch panel system integrated with the ultra-thin multi-functional film is also demonstrated, in which multi-dimensional (x-y-z) touch sensing is successfully achieved. By utilizing frequency properties of capacitive and force touch induced electric signals, these two external combined stimuli can be smoothly separated. By employing the capacitive signals, both static force touch detection and elimination of propagated stress are achieved. The presented work showcases a smart material and nano-technology combined multi-functional film, and its application as a touch panel for multi-dimensional touch sensing for smart surfaces.

Two large pieces of CVD grown mono-layer graphene[18] (5×5 cm^2) were transferred onto the surfaces of polyethylene terephthalate (PET) films (~20 μm). The relevant characterizations of the mono-layer grapheme and comparison with other works are provided in the Supporting Information. One of them is etched into four small squares (2×2 cm^2), which are used as sensing layer. The other is left intact to serve as a ground layer. The two graphene/PET samples were arranged to face each other sandwiching a piece of PVDF film (~40 μm) to form a PET/graphene/PVDF/graphene/PET multi-layer device. The graphene and PVDF form a multi-functional ultra-thin film for capacitive and force touch sensing, as conceptually shown in Figure 1 (a). The multi-layer device is then laminated to create a transparent ultra-thin multi-functional film based touch panel. The touch panel' structure and photograph are shown in Figure 1 (b) and (c), respectively.

The two graphene layers constitute a capacitor for sensing capacitive based touch events, while the PVDF layer works to detect force touch events, expanding touch detection from 2D to 3D. A conventional projected capacitance technique[5, 30] is used to measure the change of capacitance at the graphene electrodes to detect any capacitive touch event. When a conducting object (human finger or stylus) touches the screen, the electric field lines are disturbed, thus modulating the charge distribution and hence the capacitance[3]. This achieves 2D detection. Force detection in the z-direction relies on the piezoelectric output of the PVDF, which comes from its non-centrosymmetric structure[9]. When a force is applied to a PVDF film, the force induced stress gives rise to electric dipole moments, so that the polarization moves positive or negative according to the direction of the applied force[9]. The change in polarization alters the electric field inducing charge, which is then collected by a readout circuit. Since the amount of charge generated has a linear relationship with the magnitude of the applied force assuming the PVDF behaves within the elastic limit, we can calculate the magnitude of the force by measuring the amount of generated charge[8]. The generalised form relation between the mechanical and electrical domains, based on a linear approximation, can be expressed as[8]:

$$D = \varepsilon_r^{\sigma} E + d\sigma; \tag{1}$$

where σ and d denote the applied stress and the piezoelectric coefficient, respectively, E and D are the respective electric field strength and dielectric displacement, and $\boldsymbol{\varepsilon_r}$ indicates the permittivity. When the direction of the applied force is parallel to the poling direction of the PVDF film, the piezoelectric d_{33} coefficient comes into play. The scalar expressions for force touch interpretation can be expressed as:

$$\sigma = F / A; \tag{2}$$

$$P = d_{33}\sigma; \tag{3}$$

$$Q = AP = d_{33}F; \tag{4}$$

$$V = Q / C = d_{33}Ft / \varepsilon_0\varepsilon_r A; \tag{5}$$

where F and A are the applied force and the contact area, respectively, P and Q are the respective stress induced polarization and charge, d_{33} indicates the piezoelectric coefficient of the PVDF film, t is the thickness of the PVDF, and $\boldsymbol{\varepsilon_0}$ and $\boldsymbol{\varepsilon_r}$ denote the vacuum permittivity and the relative permittivity of the PVDF material, respectively. As the high $\boldsymbol{\varepsilon_r}$ of PVDF leads to current leakage, PVDF cannot measure a static force[9].

PVDF has four crystalline polymorphs: α, β, δ and γ phases[9]. Of these, the all-trans β phase was used in this work, as it has the highest piezoelectric d_{33} coefficient for forces perpendicular to the in-plane axes[9]. Its structural representation and piezoelectric principle are depicted in Fig. 1 d. As explained above, when a finger taps on the graphene/PVDF/graphene based touch panel, the generated charge can be used to interpret the force level, and the changed capacitance indicates the presence of capacitive touch, as shown in Fig. 1 a.

The experimental results below demonstrate the performance of the fabricated touch panel, in terms of sensitivity and the stability of capacitive touch and force touch detection. Gloved finger touches were carried out on the touch panel and output signal measured using a parameter analyser (Keithley 4200 SCS). The results are depicted in Fig. 2 a. Each spike indicates a touch event. The amplitude of the spikes represents the strength of the capacitive touch events. Here, strength means the amplitude of the change in capacitance, which depends on the overlapping area and the distance between the finger and the touch panel. It was observed that even a small capacitance change at 0.2 pF was successfully detected, indicating high capacitive touch sensitivity. Dynamic force touch experiments were carried out using a shaker, which outputs stable force signals at a desired amplitude and frequency. The force touch induced electric signal was sensed by a charge amplifier, and then sent to an oscilloscope. In the force touch experiment, the applied force was controlled at 1N. The results shown in Fig. 2 b show that stable force induced voltage signals of around 1.5 V (10× amplified) were obtained. The minimum detectable force strength is 0.07 N. (The comparison with relevant work is provided in Supporting Information, Table II). The mechanical stress test is performed to examine both the electrical and mechanical robustness of the sandwiched film after bending and releasing. The touch panel was bended (30 degree) up to 100 times. Quantitative capacitive and force touches

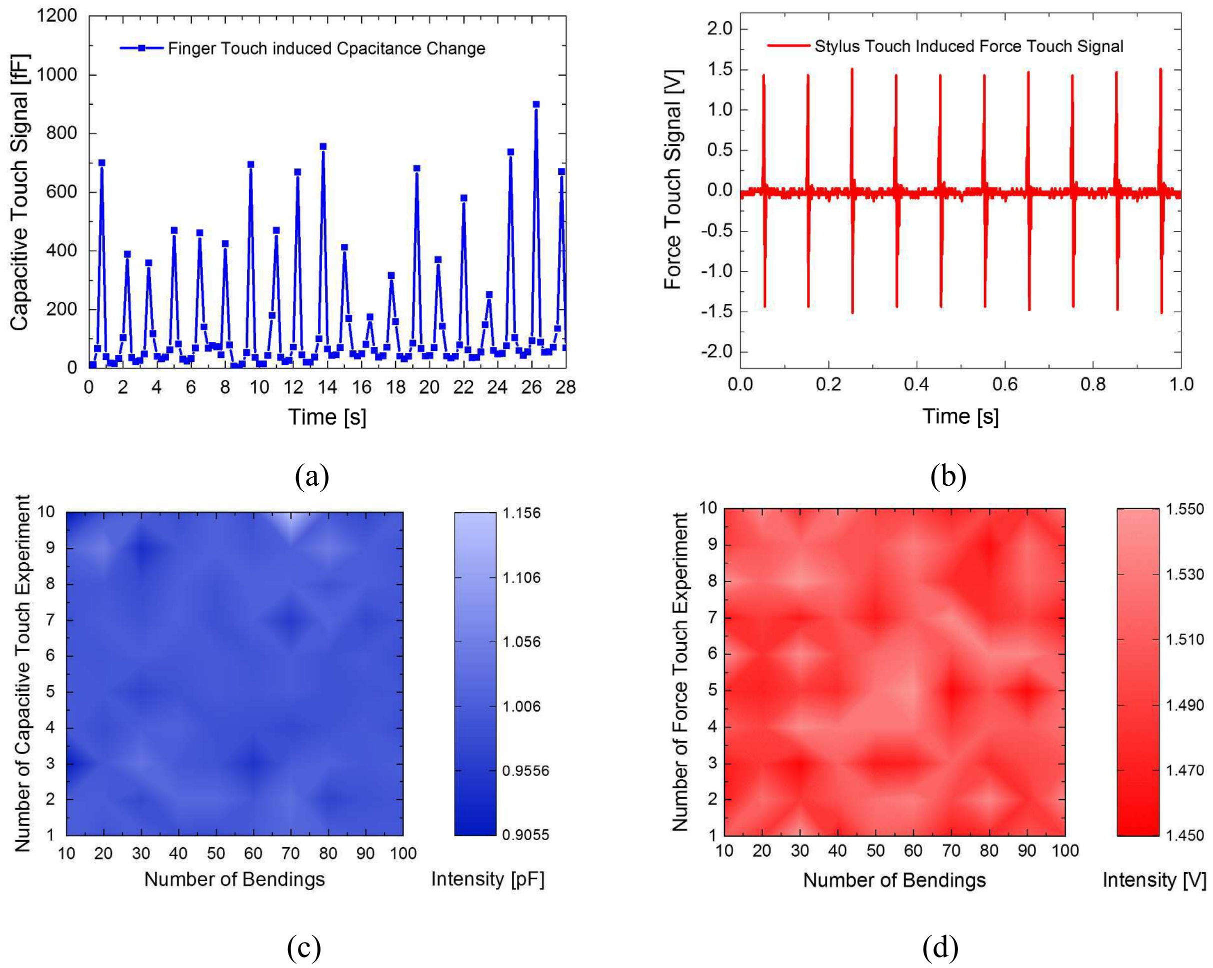

Figure 2. (a) Capacitive touches with the graphene/PVDF based multi-functional film. (b) Force touches with the graphene/PVDF based multi-functional film. (c) Stability test in terms of capacitive touch signals under different times of folding. (d) Stability test in terms of force touch signals under different times of folding. In b) and d), in each number of capacitive or force touch experiment, 10 experiments are performed and the average value is shown.

were performed after the panel had been bended every ten times. The results shown in Fig. 2 c-d confirm that the fabricated touch panel offers good electrical robustness and mechanical integrity.

To demonstrate how the simply structured multi-functional film can be used in a practical system, a touch screen panel prototype was assembled. In the system, a mutual capacitance based technique was used to detect capacitive touch events. The mutual capacitance between two graphene electrodes was measured using a high frequency electric signal (normally at 100 kHz due to the small capacitance change[3]. The equivalent circuits of the touch panel and readout circuit are depicted in Fig. 3 a and Fig. 3 b). In contrast, force detection occupies a low frequency band, as shown in Fig. 3 c. When a finger force touch is applied, the low-frequency force touch signal modulates the relatively high frequency capacitive signal, as demonstrated in Fig. 3 d. Hence, by using low-pass and band-pass filtering techniques, force touch and conventional capacitive touch events were independently retrieved.

The touch panel system is depicted in Fig. 3 e, in which a readout circuit is connected to the touch panel for signal acquisition. The readout circuit consists of a charge amplifier and an analogue to digital converter (ADC). After the ADC, the digitalized signal conveying touch information is sent to a processor. In the processor, digital low-pass and band-pass filters are applied to the retrieved touch signal, to separate capacitive and force touch events. Algorithms for different outcomes (e.g. interpreting the force level and location of the touch) computed in the processor. In this work, a laptop was used to further process the data sent from the touch panel processor. A photograph of the touch panel system is shown in Fig. 3 f.

As mentioned above, one piece of graphene was etched into four small square areas, which represent four touch pads. The main drawback of using PVDF, or any other piezoelectric material, for force sensing is its inability to detect static force[9]. Furthermore, when a force touch occurs at one location, the mechanical stress can propagate to adjacent areas, depending on the mechanical properties of the touch panel and the character of the force touch. Although the propagated stress and induced charges are small, it can become difficult to distinguish whether the signals were generated by a light touch or an adjacent heavy touch. To solve this, capacitive

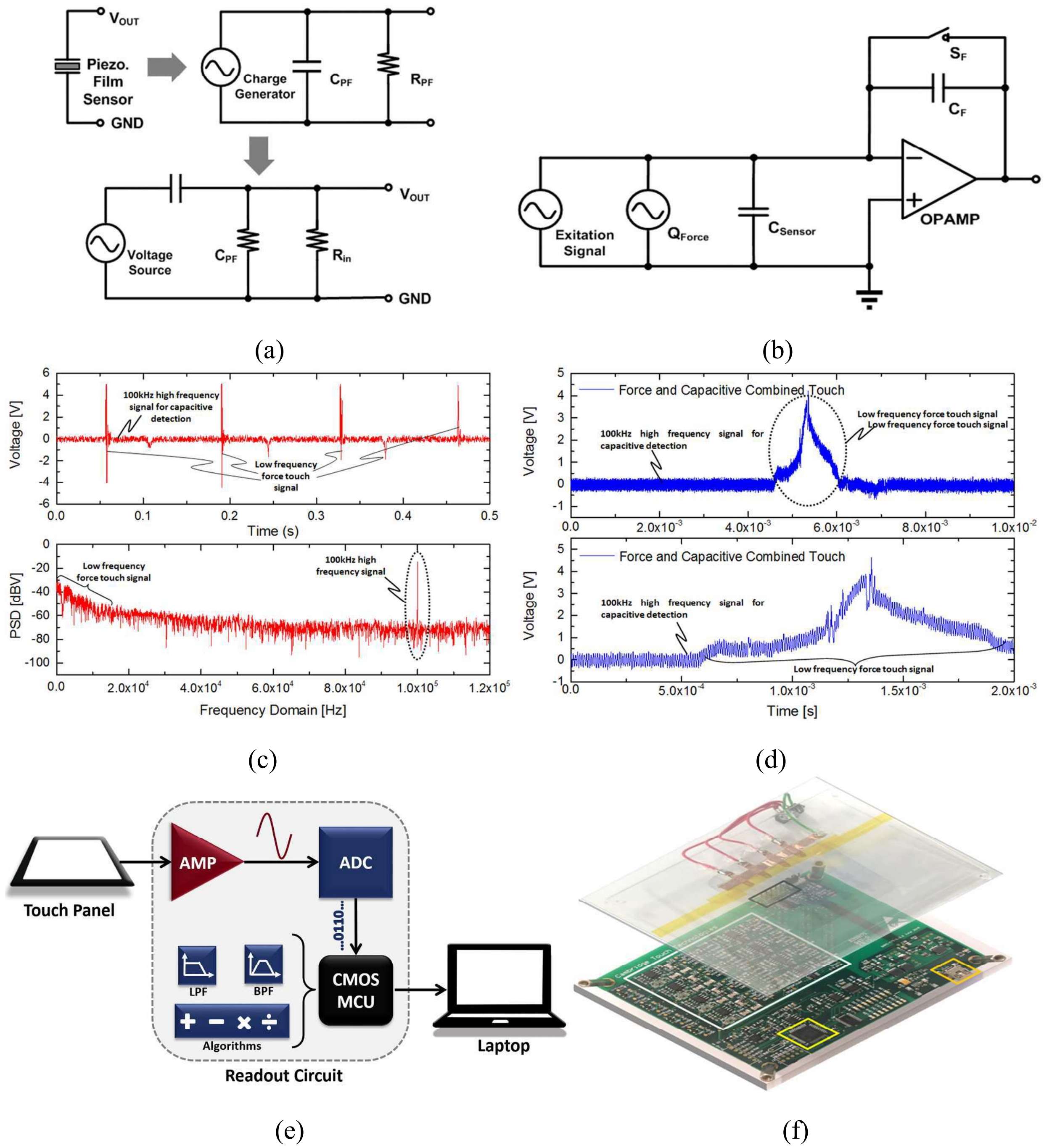

Figure 3. (a) Equivalent circuits of the piezoelectric touch panel and (b) readout circuit. Characteristics of force and capacitive touch signals in (c) frequency and (d) time domain. (e) System diagram of the touch panel system. (f) Photo of the touch panel system with the main components highlighted. White, black, yellow and orange blocks indicate charge amplifiers, touch panel interface, ADC/MCU and connection port with the laptop, respectively.

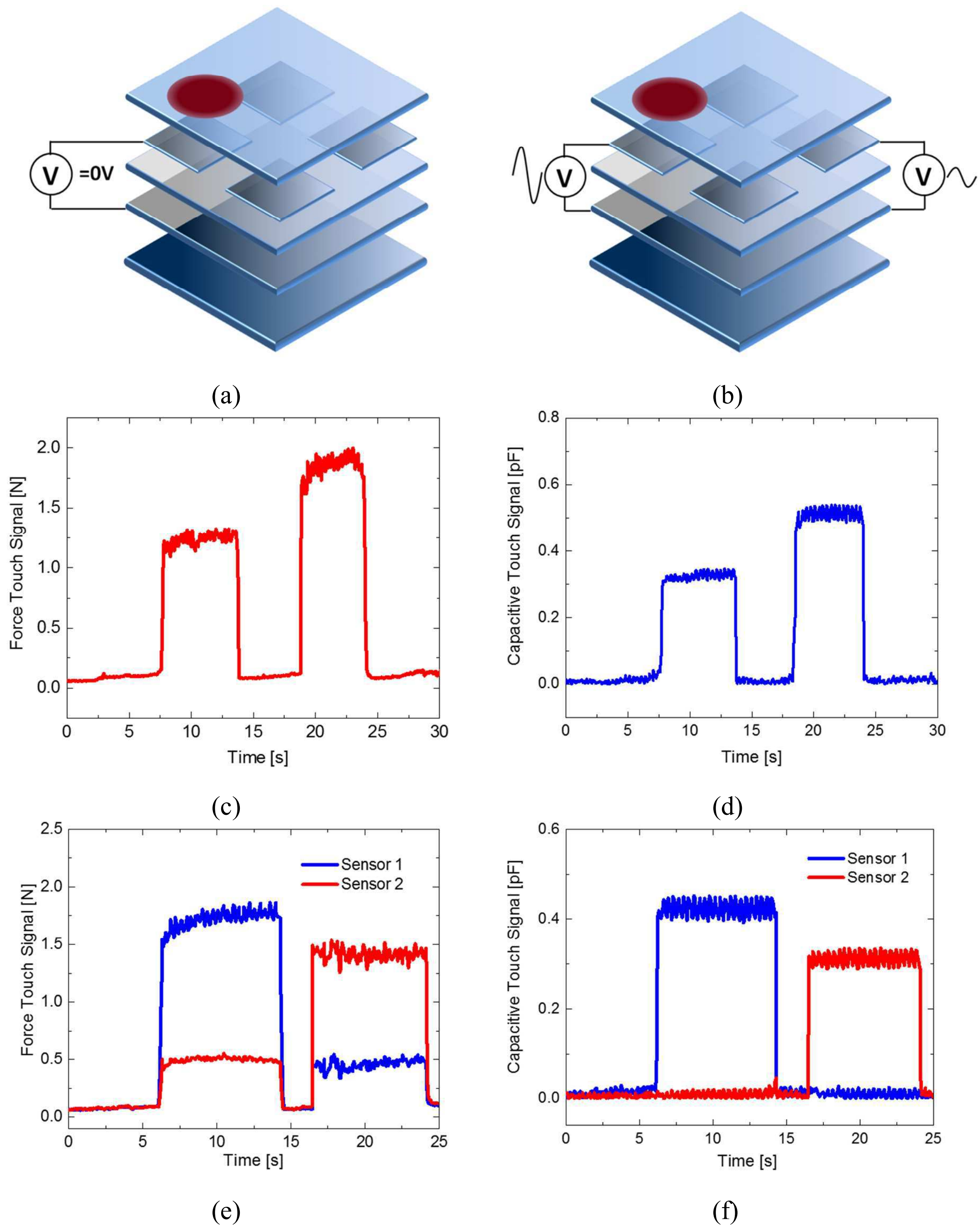

Figure 4. (a) - (b) Static force touch issue and force touch interference issue. (c) - (d) Force and capacitive signal outputs from one channel of the system. (e) - (f) Force and capacitive signal outputs from two adjacent channels of the system. It can be observed that the capacitive interference is much smaller than that of the force interference, indicating the capacitive touch signal can be used as to remove the fake force touch signals.

signals are used. The capacitive signal indicates a static force signal, as both capacitive and force signals are concurrently generated by the touch action. The corresponding experimental result is shown in Fig. 4 (a) and (b). Thus a location that experiences an adjacent force touch induced charge does not experience a change in capacitance, indicating that no touch event has occurred (as shown in Fig. 4 (c) and (d)). After applying the algorithm, the propagated stress is eliminated, which is shown in supplementary materials (ST.avi and PSE.avi).

This paper presents an ultra-thin flexible multi-functional interactive touch screen integrated with graphene and PVDF. Here, force sensing is achieved using the piezoelectric property of the PVDF thin film, thus expanding the conventional 2D (x-y) touch sensing in capacitive touch panels to 3D in which sensitivity in the z-direction provides additional functionality. Using capacitive touch signals solves the issues of static touch detection and force touch interference. The work reported in this paper not only demonstrates the integration of functional materials for advancing interactivity, but also provides insight into how the performance of piezoelectric materials is affected at the system level.

ASSOCIATED CONTENT

Supporting Information

1. Movie of multi-functional film based touch panel for static force touch sensing. (ST, avi).
2. Movie of multi-functional film based touch panel for propagated stress elimination. brief description (PSE, avi).

3. Characteristics of Graphene and compare with relevant work (Supporting Information.pdf).

AUTHOR INFORMATION

Corresponding Author

*an299@cam.ac.uk.

Author Contributions

S.G. and A.N. designed the experiments. X.W. carried the graphene work. X.W. and H. M. conducted the fabrication of the touch panel. S.G. conducted capacitive touch and force touch measurements. S.G. designed and implemented the algorithms. All authors wrote and edited the manuscript.

Notes

The authors declare no competing financial interests.

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ABBREVIATIONS

PVDF, polyvinylidene difluoride; TSP, touch screen panel; ITO, indium tin oxide; CVD, chemical vapor deposition; PET, polyethylene terephthalate; ADC, analogue to digital converter.

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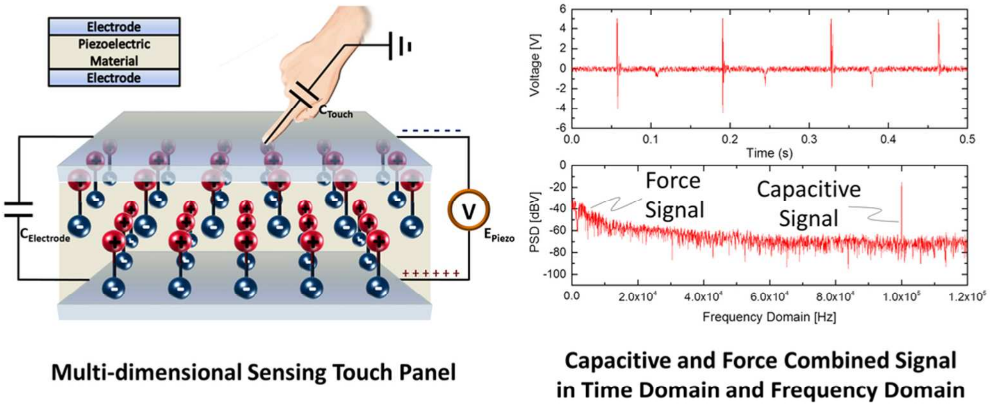

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