Graphene–ferroelectric metadevices for nonvolatile memory and reconfigurable logic-gate operations

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Memory metamaterials are artificial media that sustain transformed electromagnetic properties without persistent external stimuli. Previous memory metamaterials were realized with phase-change materials, such as vanadium dioxide or chalcogenide glasses, which exhibit memory behaviour with respect to electrically/optically induced thermal stimuli. However, they require a thermally isolated environment for longer retention or strong optical pump for phase-change. Here we demonstrate electrically programmable nonvolatile memory metadevices realised by the hybridization of graphene, a ferroelectric and meta-atoms/meta-molecules, and extend the concept further to establish reconfigurable logic-gate metadevices. For a memory metadevice having a single electrical input, amplitude, phase and even the polarization multi-states were clearly distinguishable with a retention time of over 10 years at room temperature. Furthermore, logic-gate functionalities were demonstrated with reconfigurable logic-gate metadevices having two electrical inputs, with each connected to separate ferroelectric layers that act as the multi-level controller for the doping level of the sandwiched graphene layer.

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Metamaterials are artificial media that exhibit unusual electromagnetic properties such as anomalous refraction\(^1\)–\(^3\), invisible cloak\(^5\)–\(^7\) and strong chirality\(^8\)–\(^9\). Light–matter interaction can be dramatically intensified with the use of electromagnetic responses around the resonance of specifically designed meta-atoms (MAs). Most of the wave properties, such as amplitude\(^10\), phase\(^11\) and polarization states\(^12\), and even the direction of light\(^13\), can be manipulated by the use of metamaterials or metasurfaces. Furthermore, variability in light–matter interaction is manifested through the use of metamaterials or metasurfaces. Hence, their amplitude, phase and polarization states of light can be stored to multi-states with an appropriate MA structure. Hysteretic response in the effective properties of metadevices is attributed to the change in graphene doping level (\(E_F\) of graphene in the GF-NMM as a function of \(V_G\)). Experimentally observed hysteretic variation of the spectral features is attributed to the change in graphene doping level resulting from the reversal of ferroelectric \(P\) (Supplementary Figs 2,3 and Supplementary Discussion). It is worthwhile to note that gradual transmission change is observed near positive and negative \(V_C\) and multi-state memory operation can be achieved by utilizing this gradual change near \(V_C\). To validate multi-state memory operation, a timing diagram of \(T_a\) was recorded at 0.5 THz for various \(V_G\) values of \(-80\), \(-105\), \(-115\) and \(-130\) V and plotted in Fig. 1e. From the highest \(T_A\) data states were designated as 00, 01, 10 and 11. Before addressing each state, \(V_G\) was applied to reset ferroelectric polarization to \(+P_R\). Reliable nonvolatile memory operation requires not only data separation but also long retention time. Figure 1f depicts the retention time for each stored \(T_A\) state; the GF-NMM has been observed to retain state information for all the states for over 10\(^5\) s (the standard variation for \(T_A\), \(\sigma_{state}\) is in the range between 5.62 \(\times 10^{-3}\) and 7.58 \(\times 10^{-5}\)). The extrapolated \(T_A\) for all states do not cross each other for over 8.5 \(\times 10^8\) s (\(~10\) years), which confirms that the demonstrated GF-NMM has indisputable nonvolatility. An additional measurement on THz-TDS system stability revealed that the slight variation in the \(T_A\) was mostly a result of the fluctuations of the femtosecond laser output that was used for the THz-wave generation (the fluctuation of our femtosecond laser, \(\sigma_{laser}\) was 4.43 \(\times 10^{-3}\); Supplementary Fig. 4 and Supplementary Table 1). As the MA structure used in the measurement is of a resonant type, the phase tunability and nonvolatility were also measured. In the case of phase response, \(V_G\)-dependent hysteretic behaviour and multi-state stable retention properties were found to be similar to the transmission amplitude response (Supplementary Fig. 5).

Doping concentration in graphene is more resistant to change with time when graphene–ferroelectric hybrid devices operate in the accumulation mode compared with the depletion mode, especially at the beginning of the retention measurement\(^30\). Because the graphene used in our experiments is inherently p-doped and the intrinsic doping concentration (6.17 \(\times 10^{12}\) cm\(^{-2}\)) is larger than the extrinsic doping concentration.
transmission amplitude, data states were designated as 00, 01, 10 and 11. Before each
(approximately 10 times faster switching would be
possible. Ferroelectric polarization switching depends on the
the GF-NMM is negligible, and, therefore, stable operation is
expected by a $V_{\text{G,pulse}}$ with a one and a half times greater amplitude.
concentration modulated by the ferroelectric polarization
($6.08 \times 10^{12}$ cm$^{-2}$), our device operates exclusively in the
p-doped regime (Supplementary Fig. 6 and Supplementary
Note 2). Moreover, all multi-states were addressed by applying a negative $V_{\text{G,pulse}}$ for further accumulation mode, so that the time-dependent drift in the $E_p$ of graphene in the GF-NMM is negligible, and, therefore, stable operation is possible. Ferroelectric polarization switching depends on the magnitude and duration of $V_{\text{G,pulse}}$. For an electric field of 1 MV cm$^{-1}$ corresponding to $V_{\text{G,pulse}}(+200$ V), previous studies have reported a polarization switching time of $\sim 1$ ms (refs 31,32). Approximately 10 times faster switching would be
The principle of amplitude and phase memory operation can also be applied to light polarization memory with a chiral metamaterial (chiral GF-NMM), in which the plane of linearly polarized incident light is rotated as the light travels through. Recently, it was shown that with the integration of active materials into metamaterials, polarization switching and modulation can be dynamically controlled. However, optical activity such as circular dichroism and polarization rotation in most metamaterials can so far only be tuned by continuous external optical stimuli. With the incorporation of graphene and a

Figure 1 | Graphene–ferroelectric amplitude and phase memory metadevice. (a) Schematic representation of the graphene–ferroelectric memory metadevice composed of a THz transparent electrode (TTE) with periodical metallic lines (4 μm linewidth and 2 μm spaces between the lines), ferroelectric polymer layer (2.1 μm, represented by green), single-layer graphene, hexagonal MAs and polyimide (1 μm, represented by light red) as substrate. Polarization of the incident THz is perpendicular to the TTE lines. (b) Schematic representation of the principle of nonvolatile doping in inherently p-doped graphene by ferroelectric polarization ($P$). Positive external voltage induces $+P_k$ at the surface of a ferroelectric, resulting in hole depletion in graphene while negative external voltage changes $P$ to $-P_k$, thereby resulting in the accumulation of holes in graphene. (c) Measured (open circles) and simulated (solid lines) THz transmission spectra for external pulsed gating voltage ($V_{\text{G,pulse}}$) lasting for 1 s. The red, blue and yellow lines and circles represent the results of application of $V_{\text{G,pulse}}(+200$ V), $V_{\text{G,pulse}}(-200$ V) and $V_{\text{G,pulse}}(-120$ V), respectively. (d) Hysteresis in the measured transmission amplitude ($T_A$) and the calculated Fermi level of graphene for $V_{\text{G,pulse}}$ within a range of $+200$ and $-200$ V at a specific frequency of 0.5 THz. $V_C^+$ and $V_C^-$ are the positive and negative coercive voltages, respectively. Arrow refers to the $V_{\text{G,pulse}}$ sweep direction. Logic states denoted as 00, 01, 10 and 11 correspond to the multi-level transmission amplitudes for the retention time measurement. Error bars indicate the variation of each measured logic state. (e) Timing diagram of the transmission amplitude ($T_A$) measured at 0.5 THz for various $V_{\text{G,pulse}}$ values of $-80$, $-105$, $-115$ and $-130$ V. Counting from the highest transmission amplitude, data states were designated as 00, 01, 10 and 11. Before each $V_{\text{G,pulse}}$ was applied, $V_{\text{G,pulse}}(+200$ V) was applied to reset the ferroelectric polarization to $+P_k$. (f) Transmission amplitude ($T_A$) retention time measured at 0.5 THz for $1 \times 10^5$ s and a histogram for each state.
ferroelectric into the chiral metamaterials, the polarization states of light passing through a chiral GF-NMM can be stored by $V_{G,\text{pulse}}(V)$. Strongly coupled chiral meta-molecules (MM) (ref. 34) were employed in the fabrication of the chiral GF-NMM as shown in Fig. 2a. The rest of the chiral GF-NMM is identical with the amplitude and phase GF-NMM described above (Supplementary Fig. 7 and Supplementary Note 3).

Figure 2b shows the azimuthal polarization rotation angle ($\theta$) for two distinct $V_{G,\text{pulse}}$ values. Here, $\theta$ is extracted from the phase difference between the two circular polarizations (see Methods). On applying $V_{G,\text{pulse}}(+200\text{ V})$, $\theta_{200\text{V}}$ shows a maximum value of $15^\circ$ at 1.0 THz, while $\theta_{-200\text{V}}$ exhibits a maximum value of $14^\circ$ at 0.9 THz. It can be seen from Fig. 2b that $\Delta \theta$ (=$\theta_{+200\text{V}}$ to $\theta_{-200\text{V}}$) attains the maximum value of $8^\circ$ at 1.1 THz. To trace the hysteretic behaviour in the polarization states more clearly, $\theta$ measurement was carried out at 1.1 THz and is plotted in Fig. 2c. Because of ferroelectricity, $\theta$ changes gradually near positive and negative $V_C$. Multi-level polarization states can also be stored for over $10^5$ s without much degradation as shown in the operation of three different polarization states (Supplementary Fig. 8).

Graphene–ferroelectric reconfigurable logic-gate metadevice. The underlying concept for the operation of GF-NMM can be extended to a multi-input system such as a reconfigurable logic-gate metadevice (graphene–ferroelectric reconfigurable logic-gate metadevice (GF-RLM)). For example, a two-input system can be implemented by encapsulating graphene within two controllable ferroelectric layers as shown in Fig. 3a (Supplementary Fig. 9 and Supplementary Note 4). Independent pulsed gate control ($V_{G,\text{pulse}}$) of each ferroelectric layer and the resulting combination of polarizations offered by the individual ferroelectric layers can lead to an increase in the degree of freedom in the manipulation of carrier concentration in graphene ($N_G$) when compared with the GF-NMM having a single ferroelectric layer. Corresponding to the combination of two electrical inputs (the top electrode (T) and the bottom electrode (B)), $N_G$ as well as the THz transmission through the two-input system is expected to give unique logic outputs. Figure 3b shows the combination of polarization values in the ferroelectric layers that correspond to four kinds of logic inputs, (0, 0), (0, 1), (1, 0) and (1, 1). Input logic states 1 and 0 are prepared by applying $V_{G,\text{pulse}}(+200\text{ V})$ and $V_{G,\text{pulse}}(-200\text{ V})$ to the corresponding ferroelectric layers, respectively. If graphene in GF-RLM is inherently p-doped, logic input (1, 1) depletes majority carriers (holes) and logic input (0, 0) accumulates holes in the graphene. In the two intermediate states, (0, 1) and (1, 0), the hole concentration in graphene will be set to the values that are between those corresponding to input states (0, 0) and (1, 1). The variation in $N_G$ results in a change in the transmission spectrum as shown schematically in Fig. 3c, in which the resonance frequency is red-shifted as $N_G$ increases35. With the appropriate frequency choice for data reading ($f_{\text{READ}}$), the logic output can be decoded by comparing with $T_{\text{REF1}}$ for the AND (complementary NOR) operation or $T_{\text{REF2}}$ for the OR (complementary NAND) operation. Furthermore, XOR (complementary XNOR) operation can also be realized by simply rearranging the electric connections Supplementary Figs 10,11). All the logic-gate operations are measured using THz-TDS and shown in Fig. 3d (Supplementary Fig. 12 and Supplementary Table 2).

In addition to the logic operations, the device can also be configured as a digital-to-analogue converter if the remanent polarization in each of the ferroelectric layers assumes a different value. If the two ferroelectric layers supply two different $P_k$ values, four kinds of $N_G$ levels corresponding to $(+P_{R1}+P_{R2})$, $(+P_{R1}−P_{R2})$, $(−P_{R1}+P_{R2})$ and $(−P_{R1}−P_{R2})$ are possible, which implies that $f_{\text{READ}}$ is different from $f_{\text{READ}}$ shown in Fig. 3c. The effective method to control polarization switching was demonstrated in a prior work35 in which ferroelectric switching was controlled by setting the current limitation ($I_C$, compliance current). By setting different compliance...
Figure 3 | Graphene–ferroelectric reconfigurable logic-gate metadevice. (a) Schematic representation of the graphene–ferroelectric reconfigurable logic-gate metadevice composed of a top THz transparent electrode (top; TTE, T), a ferroelectric polymer layer (2.1 μm, represented by green), a single-layer graphene, hexagonal MAs, a ferroelectric polymer layer (2.1 μm, represented by green), a bottom THz transparent electrode (bottom; TTE, B) and a polyimide layer (1 μm, represented by light red) as the substrate. Polarization of the incident THz is perpendicular to the two TTE lines. (b) Schematic representation of the four kinds of polarization alignments for input logic states. Red arrow implies an application of positive pulsed gating voltage for logic state 1 and blue arrow refers to the application of negative pulsed gating voltage for logic state 0. (c) Schematic representation of the transmission spectra for input logic states. For each input logic state, the relationship |R(0,0)|< |R(1,0)| < |R(0,1)| < |R(1,1)| is satisfied in the graphene–ferroelectric reconfigurable logic-gate metadevice because of p-doped graphene. A frequency of fREAD was designated for data reading and two reference transmission amplitudes, TREF1 and TREF2, were defined to execute AND (complementary NOR) and OR (complementary NAND) gate operations. For AND gate operation, the reference transmission amplitude is set to TREF1. For OR gate operation, the reference is TREF2. (d) Experimental transmission amplitude (TΔ) measured at 0.5 THz for the four types of logic inputs in the AND/OR gates, the XOR gate and the two-bit DAC.

Discussion
In this work, electrically programmable nonvolatile memory and reconfigurable logic-gate metadevices were demonstrated with the hybridization of graphene, a ferroelectric and MAs/molecules. These functional metadevices are the first demonstration of user-oriented general-purpose metadevice that can be configured in principle.

Methods
Fabrication processes for the GF-NMM and GF-RLM. All metallic parts of the hexagonal MAs/molecules and the TTE were made of 100-nm thick Au with a 10 nm thick Cr layer for enhanced adhesive strength. Single-layer graphene was synthesized by chemical vapour deposition on a Cu foil (G/Cu). Poly(vinylidene fluoride-trifluoroethylene), P(VDF-TrFE), manufactured by MSI Sensors Inc. was chosen as the ferroelectric polymer.

For the nonvolatile memory metadevice, a polystyrene (PI, PI-2610, HD MicroSystems) was spin-coated and cured on a Si wafer. An array of hexagonal MAs was deposited by a photolithography, thermal evaporation and lift-off process (MA/PI/Si). On a SiO2/Si wafer, a ferroelectric polymer (FP) was spin-coated, annealed at 130 °C for 1 h, and cooled down to room temperature slowly. A TTE was patterned by a photolithography, thermal evaporation of Cr/Au and lift-off process (TTE/FP/SiO2/Si). By etching the SiO2 with an HF aqueous solution, the TTE/FP/G was transferred onto graphene (G) on Cu foil. By etching the Cu foil with the Cu etchant APS-100, the TTE/FP/G was transferred onto the MA/PI/Si and thermally treated for adhesion. Finally, Si was detached mechanically. For the polarization state memory metadevice, MA/PI on Si was replaced with MM/PI, which was fabricated by stacking conjugated double Z patterns with a polyimide spacer of 2 μm.

For the reconfigurable logic-gate metadevice, an MA layer was deposited on G/Cu. An FP was spin-coated on the MA/G/Cu. In addition, a TTE as the top electrode was deposited on the FP/MA/G/Cu. By etching the Cu foil with a Cu etchant of APS-100, a TTE/FP/MA/G hybrid film was prepared. On a Si wafer sacrificial substrate, PI was spin-coated and TTE as the bottom electrode was deposited on PI/Si (TTE/PI/Si). An FP was spin-coated onto the TTE/PI/Si (FP/TTE/PI/Si). Reconfigurable logic-gate metadevice was fabricated by transferring the TTE/FP/MA/G onto the FP/TTE/PI/Si. Finally, Si was detached mechanically.

All metadevices were mounted on a punched printed circuit board for THz-TDS measurements.

THz-TDS system. To generate the terahertz signal, we used a low-temperature grown GaAs THz emitter (Tera-SED, Gigaoptics) illuminated by a femtosecond Ti:sapphire laser pulse train of wavelength 800 nm and 80 MHz repetition rate, respectively. An electro-optic sampling method was used to detect the transmitted terahertz signals in the time domain by using a (110) oriented ZnTe crystal of 1 mm thickness. The THz-TDS system has a usable bandwidth of 0.3–2.5 THz and a signal to noise ratio (S/N) of over 10,000:1.
Measurement of the chiral GF-NMM. The chiral GF-NMM was characterized using conventional THz-TDS. The metadevice was positioned between two wire-grid THz polarizers, that are mounted on motorized rotational stages with parallel or crossed configurations, to measure the co-polarized ($T_0$) and cross-polarized ($T_{\perp}$) transmission coefficients. The sample was carefully aligned to assure the TTE of the metadevice remains parallel to the front polarizer. From the measured transmission coefficients, the right and left circularly polarized transmission coefficients can be obtained as $T_{\times} = T_0 + iT_{\perp}$ and $T_{\circ} = T_0 - iT_{\perp}$. The azimuthal rotation angle can be calculated by the phase retardation between two circularly polarized waves as $\theta = \frac{1}{2}\{\text{arg}(T_{\times}) - \text{arg}(T_{\circ})\}$.

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Author contributions
W.Y.K., T.-T.K., S.H.L. and B.M. conceived the original ideas for nonvolatile memory and reconfigurable logic-gate operations. W.Y.K., H.-D.K., H.-T.K. and T.-T.K. fabricated the devices and characterized the graphene. H.-D.K., T.-T.K. and H.-S.P. performed simulations. W.Y.K., T.-T.K., H.-S.P., K.L., S.H.L. and B.M. analysed the data and discussed the results. W.Y.K., H.-D.K., T.-T.K., N.P. and B.M. wrote the paper and all authors provided feedback.

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