Vertically stacked multi-heterostructures of layered materials for logic transistors and complementary inverters

Woo Jong Yu¹, Zheng Li¹, Hailong Zhou¹, Yu Chen², Yang Wang², Yu Huang^{2,3} and Xiangfeng Duan^{1,3}*

Graphene has attracted considerable interest for future electronics, but the absence of a bandgap limits its direct applicability in transistors and logic devices. Recently, other layered materials such as molybdenum disulphide (MoS_2) have been investigated to address this challenge. Here, we report the vertical integration of multi-heterostructures of layered materials for the fabrication of a new generation of vertical field-effect transistors (VFETs) with a room temperature on-off ratio > 10³ and a high current density of up to $5,000 \text{ A cm}^{-2}$. An n-channel VFET is created by sandwiching few-layer MoS_2 as the semiconducting channel between a monolayer graphene sheet and a metal thin film. This approach offers a general strategy for the vertical integration of p- and n-channel transistors for high-performance logic applications. As an example, we demonstrate a complementary inverter with a larger-than-unity voltage gain by vertically stacking graphene, $Bi_2Sr_2Co_2O_8$ (p-channel), graphene, MoS_2 (n-channel) and a metal thin film in sequence. The ability to simultaneously achieve a high on-off ratio, a high current density and a logic function in such vertically stacked multi-heterostructures can open up possibilities for three-dimensional integration in future electronics.

avered materials such as graphene are of considerable interest as potential electronic materials for future electronics¹⁻⁸. However, the lack of a bandgap in graphene has limited the on-off current ratio of graphene-based transistors for logic applications. The approaches to address this challenge include the induction of a transport gap in graphene nanostructures9-13 or bilayer graphene^{14–19}, and the design of innovative device architectures such as vertical tunnelling transistors^{20,21} and barristors²². These approaches have proved successful in improving the onoff ratio of the resulting devices, but often at a severe sacrifice of the deliverable current density. Here we report the vertical integration of multi-heterostructures of layered materials (for example, graphene, MoS₂ or cobaltites Bi₂Sr₂Co₂O₈) to enable high-current-density VFETs. We show that an n-channel VFET with a room-temperature on-off ratio $>10^3$ can be created by vertically sandwiching semiconducting few-layer MoS₂ between a monolayer graphene sheet and a metal thin film. Importantly, with an ohmic-contacted metal top-electrode and an ultrathin lavered MoS₂ semiconductor channel, the VFET can deliver a high current density of 5,000 A cm⁻², about 2–5 orders of magnitude larger than the recently reported vertical tunnelling transistors or barristors^{20,22}, while retaining a high on-off ratio. Taking a step further, we demonstrate that a complementary inverter with voltage gain can be created by vertically stacking the layered materials of graphene, Bi₂Sr₂Co₂O₈ (p-channel), graphene, MoS₂ (n-channel) and a metal thin film in sequence. Our study demonstrates a general strategy for the integration of various layered materials to achieve functional circuits in the vertical direction, and can open up a new dimension for future electronics to enable three-dimensional integration.

The fabrication procedures for the VFET are schematically illustrated in Supplementary Fig. S1. In brief, monolayer graphene

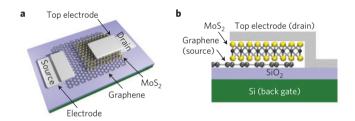


Figure 1 | Schematic illustration of the vertically stacked graphene-MoS₂-metal FETs. **a**, A schematic illustration of the three-dimensional view of the device layout. **b**, A schematic illustration of the cross-sectional view of the device, with the graphene and top metal thin-film functioning as the source and drain electrodes, and the MoS₂ layer as the vertically stacked semiconducting channel, with its thickness defining the channel length. A silicon back gate is used with a 300 nm SiO₂ dielectric layer.

was first grown by the chemical vapour deposition approach and transferred onto a silicon wafer with 300 nm SiO₂ (ref. 23). The graphene was patterned into strips of 10 μ m width and 50 μ m length by oxygen plasma etching through a photo-resist mask for a bottom electrode. Micromechanical cleavage²⁴ was used to exfoliate few-layer MoS₂ onto the patterned graphene as a semiconductor material. The top metal electrode was patterned on the MoS₂ to overlap with the bottom graphene electrode by electron-beam lithography and electron-beam deposition of Ti/Au (50/50 nm). The current flows between the bottom graphene electrode and the top metal electrode through the semiconducting MoS₂ channel, which is modulated by the silicon back gate (Fig. 1a,b). Owing to the finite density of states and the weak electrostatic screening effect

¹Department of Chemistry and Biochemistry, University of California, Los Angeles, California 90095, USA, ²Department of Materials Science and Engineering, University of California, Los Angeles, California 90095, USA, ³California Nanosystems Institute, University of California, Los Angeles, California 90095, USA, ³California 90095, USA, *e-mail: xduan@chem.ucla.edu.

ARTICLES

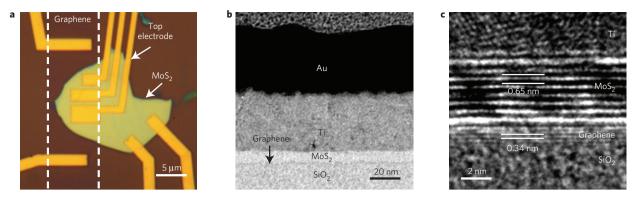


Figure 2 | **Fabrication and structural characterization of the vertical transistor. a**, An optical image of a typical vertical transistor device. The bottom graphene electrode is shaped to strips of 10 μ m in width. The multilayer MoS₂ is located on the graphene electrode and the top metal electrode is located on the MoS₂ to overlap with the bottom graphene electrode to enable vertical current flow. **b**, A cross-sectional TEM image of a vertical transistor. **c**, A cross-sectional high-resolution TEM image of the interface between the multilayer graphene, the multilayer MoS₂ and the top electrode of Ti. A TEM image of a multilayer graphene device is shown here for better illustration because it is difficult to visualize the monolayer graphene electrode owing to severe electron-beam damage while conducting the TEM studies.

of the monolayer graphene sheet, the applied back gate electric field can effectively penetrate through the graphene to modulate the energy band of the MoS_2 .

Figure 2a shows an optical image of our typical VFETs. The 10 µm strip of monolaver graphene is located inside the dotted lines. A MoS₂ flake and three top metal electrodes of variable areas overlap on top of the graphene. The thickness of the MoS₂ flake was determined by atomic force microscope (AFM) analysis to be 30 nm (Supplementary Fig. S2). The total channel area is defined by the overlapping area of the graphene and the top metal electrode. Two separate pairs of electrodes were formed on graphene and MoS₂ in the non-overlapping region to characterize the planar electrical performance. Cross-sectional transmission electron microscope (TEM) analysis was used to study the overall integration of the graphene-MoS₂-top-electrode vertical stack. The complete stack of SiO₂-graphene-MoS₂-Ti-Au can be readily seen in the low-magnification cross-section TEM image (Fig. 2b). A high-resolution TEM image of the SiO₂-graphene-MoS₂-Ti interface shows that the graphene layers are intimately integrated with the MoS₂ layers without any obvious gap or impurities between them (Fig. 2c). The layer spacings are 0.65 nm and 0.34 nm in the MoS₂ layer and the graphene layer, respectively. A TEM image of a multilayer graphene device is shown here because of the difficulties in visualizing a monolayer graphene device due to the severe electron-beam damage while conducting TEM studies. Together, these studies clearly demonstrate that the physical assembly approach can effectively integrate layered graphene and MoS₂ to form intimate contact.

Electrical transport studies of the vertical transistors and planar transistors were carried out under ambient conditions at room temperature. We first measured the output characteristics of the vertical transistor (Fig. 3a). The vertical transistor in this study has a channel length (MoS₂ thickness) of 36 nm as determined by AFM measurement. The current was normalized by the overlapping area of the graphene and the top metal electrode to obtain the current density. The output characteristics at various back-gate voltages show clearly that the current density decreases with increasing negative gate potential, demonstrating that the electrons are the majority charge carriers in this vertical transistor, which is consistent with the typical n-type semiconducting characteristics observed in MoS₂ materials previously^{25,26}. In the negative sourcedrain voltage (V_{sd}) regime, clear on and off current modulation can be achieved by varying the gate bias. On the other hand, in the positive $V_{\rm sd}$ regime, a much smaller gate modulation is observed. This difference can be attributed to the asymmetrical contact at the source and drain end, and will be further discussed in Fig. 4.

Figure 3b shows the transfer characteristics $(I_{sd}-V_g \text{ curves})$ for the same device at $V_{sd} = -0.1$, -0.2 and -0.5 V. Overall, the device shows a room-temperature on-off current ratio of \sim 1,500 at all V_{sd} values, which is about 1-2 orders of magnitude better than typical graphene devices at room temperature, and is already sufficient for typical logic device applications. The device delivers a large on-current density of 2,600 A cm⁻² at $V_{sd} = -0.5$ V and $V_g = 60$ V. In general, the maximum on-current density in an optimized VFET device can readily exceed 5,000 A cm⁻² at $V_{sd} = -1.0$ V. It is particularly important to note that this current density is about 3-5 orders of magnitude larger than the recently reported vertical tunnelling transistors²⁰ and barristor²² at the same source-drain bias. A higher current density has been achieved in a tunnelling device with ultrathin boron nitride tunnelling barriers (for example, 1–3 layers)²¹ and in a barristor with a larger bias²², but typically with rather weak gate modulation. With our VFET structure, a high current density and a high on-off ratio are simultaneously achieved for the first time, which is essential for high-performance logic transistors. Furthermore, the maximum on-current density in our VFET device can readily exceed 350,000 A cm⁻² at $V_{\rm sd} = -4$ V without consideration of the on-off ratio (Supplementary Fig. S3), which is about two orders of magnitude larger than the maximum current density of 5,000 A cm⁻² at $V_{sd} = 4$ V of the recently reported barristors.

To further confirm the vertical charge transport in the VFETs, the electrical characteristics of the planar transistors with few-layer MoS₂ as the planar channel are characterized as a control. Figure 3c shows the output characteristics of the planar transistor with nonoverlapping bottom graphene and top metal source-drain electrodes, and Fig. 3d shows the output characteristics of planar transistors between two top metal source-drain electrodes, as shown schematically in the insets of the respective figures. In these output characteristics, the current is normalized by the channel width. Although it is not necessarily accurate to compare area current density versus line current density, it is still important to note that the actual current of our VFETs is at least more than 20 times larger than those of planar transistors with similar dimensions. This striking difference in current amplitude between the vertical and planar devices confirms that the charge transport in the vertical direction indeed dominates the vertically stacked devices. Furthermore, studies on vertical devices with variable area in the range of $6-15 \,\mu\text{m}^2$ result in comparable normalized area current densities (Supplementary Fig. S4), further demonstrating that the vertical charge transport is the dominant mechanism for the current flow in the vertical devices.

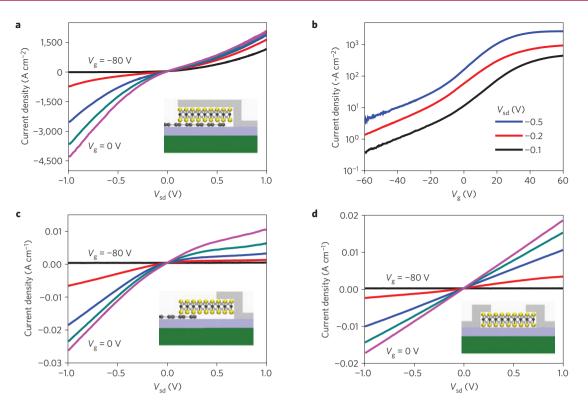


Figure 3 Room-temperature electrical properties of the vertical and planar transistors. a, $I_{sd}-V_{sd}$ output characteristics of a vertical transistor. The current is normalized by the area. **b**, $I_{sd}-V_g$ transfer characteristics of the device shown in **a** at $V_{sd} = -0.1$, -0.2 and -0.5 V. **c**, $I_{sd}-V_{sd}$ output characteristics of a planar transistor with a MoS₂ channel between a graphene electrode and a metal electrode. **d**, $I_{sd}-V_{sd}$ output characteristics of a planar transistor with a MoS₂ channel. The current is normalized by the width of the electrode in **c**,**d**. The back-gate voltage is varied from -80 (black) to 0 V (magenta) in steps of 20 V in **a**,**c**,**d**. Insets in **a**,**c**,**d** show schematics of the transistor structure for each of the output characteristics. Green, silicon substrate; blue, silicon oxide; black, graphene; yellow, MoS₂; grey, Ti (see Fig. 1b).

Figure 3a shows that the vertical transistor exhibits directionally dependent current flow and asymmetrical gate modulation, in which the output characteristics of the VFET were different under negative and positive source-drain biases. Clearly, under negative $V_{\rm sd}$, the device has a large gate modulation exceeding three orders of magnitude, whereas under positive V_{sd}, the device shows a much smaller gate modulation. These results can be explained by using the band diagrams shown in Fig. 4b,c, depicting the cases of negative and positive V_{sd}, respectively. The gate electric field is applied between the silicon back gate and the grounded top metal electrode. The graphene-MoS₂ barrier height and the MoS₂ Fermi level can be effectively modulated near the graphene-MoS₂ contact (Fig. 4a) because of the limited density of states and the weak electrostatic screening effect of graphene (Fig. 4b). In contrast, the MoS₂ Fermi level near the MoS₂-metal contact is effectively pinned and can barely be modulated by the gate potential owing to the strong screening effect of the top metal electrode (Fig. 4c). As a result, an asymmetric gate modulation is expected between positive and negative $V_{\rm sd}$.

The application of a negative V_{sd} across the device makes electrons flow from the graphene to the top electrode. In this case, the Schottky barrier at the contact between graphene–MoS₂ plays a dominant role in current modulation (Fig. 4b), similar to the recently reported barristors²². A positive gate voltage effectively reduces the graphene–MoS₂ Schottky barrier height as well as the depletion width in MoS₂ layer, allowing electrons to readily overcome the Schottky barrier through thermionic emission or thermionic field emission processes, and therefore enhances the charge transport characteristics of the vertical device. In contrast, a negative gate voltage increases the Schottky barrier height and depletion width and suppresses the electron transport across the vertical stack. In this way, the device essentially operates as an n-type field-effect transistor (FET) in the negative V_{sd} regime where the application of the gate voltage can effectively modulate the charge transport across the MoS₂ layer to result in a large on–off ratio in the vertical devices. On the other hand, under positive V_{sd} conditions across the device, electrons are injected into MoS₂ through the top metal electrode and roll downhill in MoS₂ to the bottom graphene electrodes. Therefore, the top metal–MoS₂ contact plays the primary role in determining the current modulation (Fig. 4c). With the electrostatic screening effect of the top metal electrode, the weak gate modulation on the barrier at the top electrode–MoS₂ contact thus leads to a much smaller on–off ratio.

To further probe the charge transport through graphene– MoS_2 – metal VFETs, we have carried out temperature-dependent studies. A series of I_{sd} – V_g plots ($V_{sd} = -0.1$ V) obtained at different temperatures demonstrates that the on-current exhibits a relatively small temperature dependence whereas the off-current decreases exponentially with decreasing temperature, with the on–off ratio increasing from 60 at 290 K to 15,000 at 150 K (Fig. 4d). The variable-temperature transport measurements allow us to determine the Schottky barrier heights across the graphene– MoS_2 junction. According to the thermionic emission theory, the diode saturation current is related to the Schottky barrier height by the following equation:

$$I_{\text{sat}} = AA^*T^2 \exp\left(-\frac{q\varphi_{\text{B}}}{k_{\text{B}}T}\right)$$
(1)

where A is the area of the Schottky junction, $A^* = 4\pi q m^* k_B^2 h^{-3}$ is the effective Richardson constant, q is the elementary charge, k_B is

NATURE MATERIALS DOI: 10.1038/NMAT3518

ARTICLES

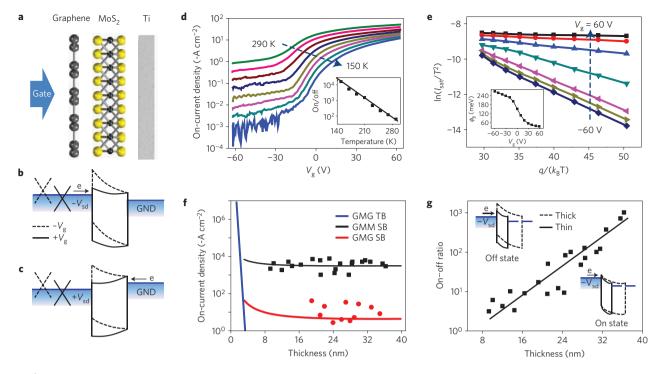


Figure 4 | **Schematic illustration of the band diagrams of the vertical transistors and the electrical characteristics. a**, Schematic illustration of a vertical transistor. Gate electric field is applied from silicon back gate and the top metal electrode is connected to ground (GND). **b**,**c**, The band structure at negative source bias at graphene (**b**, $V_{sd} < 0$) and positive source bias at graphene (**c**, $V_{sd} > 0$) with the top metal electrode connected to ground under positive (solid) or negative (dashed) V_g . **d**, Transfer characteristics of a VFET at different temperatures from 290 to 150 K ($V_{sd} = -0.1$ V). The inset shows the corresponding on-off ratio variation. **e**, Temperature-dependent diode characteristics. Each curve from the bottom to the top is obtained at different V_g values from -60 to 60 V with 20 V step variation. The inset shows the corresponding Schottky barrier height variation obtained from the slope of the fitted line. **f**, The simulated on-current density of the GMG TB (blue line), graphene-MoS₂-metal (Ti) Schottky barrier (GMM SB, black line) and GMG SB (red line) configurations at a bias of -0.1 V. Experimental on-current density is plotted as black squares for GMM SB and red circles for GMG SB at $V_{sd} = -0.5$ V. **g**, The on-off current ratio of the vertical transistors with various channel lengths (thicknesses of MoS₂). The on- and off-state band diagrams for thin (solid) and thick (dashed) MoS₂ layers are represented in the insets.

the Boltzmann constant, T is the temperature, m^* is the effective mass and h is the Plank constant²². The saturation current was determined from a logarithmic plot of the forward-bias current (Supplementary Fig. S5). The ideality factor of the graphene–MoS₂ diode can also be determined from this plot to be 3-4.5. The deviation from an ideal diode may be attributed to graphene-MoS₂ interface defects and also probably the electrostatic screening effect of the top MoS₂-metal contact due to the very small MoS₂ channel thickness (length). On the basis of the relation in equation (1), the Schottky barrier height can be estimated from the slope of the $\ln(I_{\text{sat}}/T^2)$ versus $q/k_{\text{B}}T$ plots (Fig. 4e). The derived Schottky barrier shows a clear dependence on the gate voltage (V_g) : changing from 260 to 33 meV as the V_g is increased from -60 to 60 V. This change in Schottky barrier height can be largely attributed to modulation of the graphene work function²². It is also important to note that the Schottky barrier height is hardly changed at large negative and large positive gate voltage points when the Fermi level $(E_{\rm F})$ of graphene is located far from the Dirac point because of the relatively high density of states at these points. However, the Schottky barrier height is markedly changed near 0V gate bias when $E_{\rm F}$ is located near the Dirac point in graphene because of the much lower density of states. The Schottky barrier height at $V_{\rm g} = 0 \,\mathrm{V}$ is 150 meV, which is much smaller than the 400 meV value expected from the energy level difference between the work function of graphene (4.5 eV) and the electron affinity of MoS₂ (~4.1 eV). This difference may be attributed to the electrostatic screening effect of the top metal electrode that could effectively reduce the bottom Schottky barrier height (and width) owing to the ultrashort channel length ($\sim 20 \text{ nm}$).

It is important to note that our VFET is fundamentally different from the recently reported vertical tunnelling transistors in that the MoS₂ layer in our device functions as a true semiconducting channel instead of an insulating tunnelling barrier²⁰, which has allowed us to achieve a much larger vertical current flow while retaining a high on-off ratio. In general, our device exhibits a unipolar behaviour with electrons as the majority carriers (dictated by the n-type MoS₂ layer) instead of bipolar characteristics in vertical tunnelling transistors with the carrier type determined by the gate-modulated graphene. To further understand the difference between our VFETs and the reported vertical tunnelling transistors, we have considered three different device configurations including a graphene-MoS₂-graphene tunnelling barrier (GMG TB; Supplementary Fig. S6a), a graphene-MoS2-graphene Schottky barrier (GMG SB; Supplementary Fig. S6b) and a graphene–MoS₂–metal (Ti) Schottky barrier (GMM SB; Supplementary Fig. S6c) with variable MoS₂ thickness and calculated the corresponding current densities for each configuration (see Supplementary Information for details).

For the GMG stack with a very thin MoS_2 layer (<4 nm), the Fermi level in MoS_2 is effectively pinned by both the bottom and top junctions and can barely be modulated by the back gate (Supplementary Fig. S6a). The tunnelling current is the dominant component in charge transport (blue line in Fig. 4f and Supplementary Fig. S7). Our calculation shows that a high current density (>10¹ A cm⁻²) can be achieved by using 1–4 layers of MoS_2 tunnelling barrier but typically with a rather weak gate modulation (<10²); and a rather large on–off current ratio can be achieved by using 5–7 layers of MoS_2 (~2.5–4 nm) tunnelling barrier, but with

ARTICLES

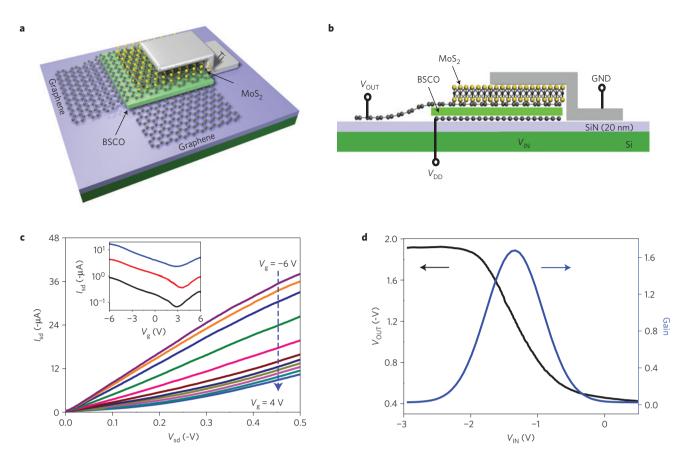


Figure 5 | **Vertically stacked multi-heterostructures of layered materials for complementary inverters. a**, Three-dimensional schematic illustration of a complementary inverter by vertically stacking the layered materials of graphene, $Bi_2Sr_2Co_2O_8$ (p-channel), graphene, MoS_2 (n-channel) and a metal thin film on a Si/SiN_x (20 nm) substrate. **b**, Cross-sectional view of the vertically stacked inverter, with the bottom graphene as the source terminal (V_{DD}), the top metal electrode as the ground (GND), the intermediate graphene as the output (V_{OUT}) and the bottom silicon as the input (V_{IN}). **c**, Output characteristics of a p-channel $Bi_2Sr_2Co_2O_8$ VFET with top and bottom graphene electrodes on a Si/SiN_x (20 nm) substrate. The back-gate voltage is varied from -6 V (top) to 4 V (bottom) in steps of 1V. The inset shows the transfer characteristics of $Bi_2Sr_2Co_2O_8$ VFET at $V_{sd} = -0.2, -0.05$ and -0.01 V from top to bottom. **d**, The inverter characteristics from vertically stacked p- and n-type VFETs. A negative supply voltage ($V_{DD} = -2 V$) is applied to the bottom graphene, and the gain of the inverter is ~ 1.7 .

a relatively small current density ($<1 \text{ A cm}^{-2}$; see Supplementary Fig. S7, as also confirmed by ref. 20). For even thicker MoS₂ layers (>4 nm), the thermionic emission through the Schottky barrier becomes the dominant component in charge transport. The calculated thermionic emission current shows a rather weak dependence on the MoS₂ thickness in the range of 4–40 nm, probably due to a complete depletion of the semiconductor layer and downhill charge transport after crossing the barrier. It is important to note that there is a substantial difference between the GMM (Ti) SB and the GMG SB device: the calculated current density of GMM (Ti) SB (black line in Fig. 4f) is about three orders of magnitude larger than that of the GMG SB configuration (red line in Fig. 4f).

We have experimentally investigated a large number of GMM (Ti) SB devices with variable MoS_2 thickness and device size. The overall on-current density is typically in the range of $10^3 \sim 10^4$ A cm⁻² with a weak dependence on the MoS_2 layer thickness (black squares in Fig. 4f), matching well with the calculated current density. Studies on GMG SB devices with variable MoS_2 thickness have also been carried out as a control experiment (Supplementary Fig. S8). The obtained on-current in GMG SB devices (red dots in Fig. 4f) is about 2–3 orders of magnitude smaller than that of GMM (Ti) SB devices, consistent with theoretical calculations. This consistent difference observed in theoretical modelling as well as experimental studies clearly demonstrates that our GMM SB device is fundamentally distinct from the GMG SB structure.

In the GMG SB with a thick MoS₂ layer, two Schottky barriers are formed at both MoS₂-graphene contacts with the bottom one strongly modulated and the top one weakly modulated by the back gate in the opposite direction (Supplementary Fig. S6b). Even though the electrons can be effectively injected through the bottom barrier at the on-state, the top Schottky barrier can suppress the overall current flow to result in a lower current density. In contrast, for the GMM SB configuration, the top Schottky barrier height is minimized and can hardly be modulated by using a low-workfunction metal (Ti) to make a nearly ohmic contact with MoS₂ (Supplementary Fig. S6c). In this way, the charge transport is dominated by the bottom graphene-MoS₂ Schottky barrier height and width, which can be effectively modulated by the back-gate voltage to allow for highly efficient injection and transportation of the electrons through the MoS₂ layer, enabling a very large current density that is about three orders of magnitude larger than that of the GMG TB or GMG SB devices with similar on-off ratios. Furthermore, comparing the recently reported barristors having bulk silicon as the semiconductor layer with our GMM SB device, the charge transport through thick bulk silicon ($\gg 10 \,\mu m$ Si) and the thick depletion layer at the graphene-Si junction can a severely limit the overall current flow to result in a current density that is also about 2-3 orders of magnitude smaller than our GMM SB VFETs reported here²².

Figure 4g shows the room-temperature on-off current ratios of the VFETs. The on-off ratios are strongly dependent on the

MoS₂ thickness. In general, it is found that the room-temperature on-off ratio of the vertical transistors can exceed three orders of magnitude with a relatively thick MoS₂ layer (for example, 30–40 nm), and gradually decreases to 3 when the MoS₂ thickness is reduced to 9 nm or so. This trend can be explained by a shortchannel effect. With decreasing MoS₂ thickness, the potential of the entire channel (including the graphene–MoS₂ contact) becomes more and more dominated by the electric field of the top metal electrodes, which can reduce the bottom off-state Schottky barrier height and width to result in a smaller on-off ratio (Fig. 4g, insets). The room-temperature on-off ratio of $>10^3$ achieved in our devices is about two order of magnitude smaller than the best on-off ratio achieved in recently reported barristors²², which might be attributed to the presence of interface defects at the graphene–MoS₂ interface, and can be improved to $>10^4$ on cooling (as demonstrated by our low-temperature studies, Fig. 4b) or with cleaner interfaces in future studies.

This strategy of vertical integration is general and can be readily extended to various layered materials to obtain vertically stacked devices with both n- and p-channel characteristics. For example, layered transition-metal oxides such as cobaltites Bi2Sr2Co2O8 (BSCO; refs 27,28) can be exfoliated into single- or few-layer materials and exhibit p-channel characteristics (Supplementary Fig. S9). Taking a step further, multiple layers of layered materials can be vertically stacked to enable more complicated device functions. To this end, we have demonstrated a complementary inverter by vertically stacking multi-heterostructures of the lavered materials including graphene, BSCO (p-channel), graphene, MoS₂ (n-channel) and a metal thin film on a Si/SiN_r (20 nm) substrate (Fig. 5a,b, and Supplementary Fig. S10). In this complementary inverter, the vertically stacked BSCO layer functions as a p-channel VFET, and the MoS₂ layer functions as the n-channel VFET. A supply voltage was applied to the bottom layer of graphene and the top metal electrode was connected to the ground. The output voltage was measured on the intermediate layer of graphene as a function of the input voltage on the silicon back gate. Electrical measurement shows that the vertically stacked BSCO layer with both bottom and top graphene contacts shows clear p-channel characteristics (Fig. 5c). More importantly, the top stack of n-channel MoS₂ VFET retains excellent switching characteristics (Supplementary Fig. S11), demonstrating that the bottom-gate electrical field can readily penetrate through the entire bottom p-channel device to effectively modulate the top n-channel VFET. In this way, the vertically stacked multi-heterostructures forms a complementary inverter with a larger-than-unity voltage gain (1.7; Fig. 5d).

We have demonstrated that vertical integration of heterostructures of layered materials can enable a new design of graphene-based transistors simultaneously with both a high on-off current ratio and a high current density at room temperature, to satisfy two critical requirements for high-performance logic applications. With the use of an ultrathin layered semiconductor channel and an ohmic-contacted top metal electrode, our VFETs can deliver an unprecedented current density that is 2-5 orders of magnitude larger than that of the recently reported graphene-based vertical tunnelling transistors and barristors^{20,22}, while retaining a roomtemperature on-off current ratio exceeding 3 orders of magnitude. It should be noted that the achievement of a high current density is central to the performance of a transistor because the intrinsic delay of a transistor ($\tau = CV/I$) is inversely proportional to the deliverable current density. With a similar device geometry, a higher current at the same bias can be directly translated into a higher device speed. Taking a step further, we have also demonstrated that the vertical integration of multi-heterostructures of layered materials can be used to effectively create more complicated device functions including a complementary inverter with a larger-thanunity voltage gain. A striking point that we demonstrate here is that the electrical field can readily penetrate through the first layer of p-FET to effectively modulate the second layer n-FET, making the vertical integration of multi-heterostructures possible and meaningful for functional devices. Our study demonstrates the feasibility to implement vertically stacked devices for useful logic functions with gain. This is fundamentally different from the conventional planar electronics and the recently reported logic functions achieved through the lateral integration of p- and n-type barristors²². Our study demonstrates a general strategy to integrate layered materials in the vertical direction to enable functional devices and circuits, and can open up a new dimension for high-density integration of functional devices in the limited circuit area.

Methods

Synthesis and characterization. The graphene was grown by chemical vapour deposition on copper foil at 1,050 °C with methane as the carbon-containing precursor. After growth, the graphene was transferred onto a silicon/silicon oxide substrate and patterned into 50 by 10 μ m strips by oxygen plasma etching. Molybdenum disulphide flakes (MoS₂) were then exfoliated onto the graphene strips using a micromechanical cleavage approach. The top metal electrode was patterned on the MoS₂ to overlap with the graphene by electron-beam lithography and electron-beam deposition of Ti/Au (50/50 nm).

Microscopic and electrical characterization. The cross-sectional TEM sample was prepared by focused ion beam cutting and was characterized by a TF20 TEM operating at 300 kV. Tapping-mode AFM was carried out with a Veeco 5000 system. Scanning electron microscope imaging was performed on a JEOL 6700F unit operated at 5 kV. The d.c. electrical transport studies were conducted with a probe station at room temperature under ambient conditions with a computer-controlled analog-to-digital converter.

Received 2 July 2012; accepted 6 November 2012; published online 16 December 2012

References

- Novoselov, K. S. *et al.* Electric field effect in atomically thin carbon films. *Science* 306, 666–669 (2004).
- 2. Novoselov, K. S. et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 438, 197–200 (2005).
- Dean, C. R. et al. Boron nitride substrates for high-quality graphene electronics. Nature Nanotech. 5, 722–726 (2010).
- 4. Wu, Y. Q. *et al.* High-frequency, scaled graphene transistors on diamond-like carbon. *Nature* **472**, 74–78 (2011).
- Liao, L. *et al.* High-speed graphene transistors with a self-aligned nanowire gate. *Nature* 467, 305–308 (2010).
- 6. Schwierz, F. Graphene transistors. *Nature Nanotech.* 5, 487–496 (2010).
- 7. Wang, F. *et al.* Gate-variable optical transitions in graphene. *Science* **320**, 206–209 (2008).
- Weiss, N. O. et al. Graphene: An emerging electronic material. Adv. Mater. 24, 5782–5825 (2012).
- Han, M. Y., Ozyilmaz, B., Zhang, Y. & Kim, P. Energy band-gap engineering of graphene nanoribbons. *Phys. Rev. Lett.* 98, 206805 (2007).
- Li, X., Wang, X., Zhang, L., Lee, S. & Dai, H. Chemically derived, ultrasmooth graphene nanoribbon semiconductors. *Science* 319, 1229–1232 (2008).
- 11. Bai, J., Duan, X. & Huang, Y. Rational fabrication of graphene nanoribbons Using a nanowire etch mask. *Nano Lett.* **9**, 2083–2087 (2009).
- Wang, X. & Dai, H. Etching and narrowing of graphene from the edges. Nature Chem. 2, 661–665 (2010).
- Bai, J., Zhong, X., Jiang, S., Huang, Y. & Duan, X. Graphene nanomesh. *Nature Nanotech.* 5, 190–194 (2010).
- 14. Zhang, Y. *et al.* Direct observation of a widely tunable bandgap in bilayer graphene. *Nature* **459**, 820–823 (2009).
- Oostinga, J. B. *et al.* Gate-induced insulating state in bilayer graphene devices. *Nature Mater.* 7, 151–157 (2008).
- Ohta, T., Bostwick, A., Seyller, Th., Horn, K. & Rotenberg, E. Controlling the electronic structure of bilayer graphene. *Science* 313, 951–954 (2006).
- Xia, F., Farmer, D. B., Lin, Y. & Avouris, Ph. Graphene field-effect transistors with high on–off current ratio and large transport bandgap at room temperature. *Nano Lett.* **10**, 715–718 (2010).
- Yu, W. J., Liao, L., Chae, S. H., Lee, Y. H. & Duan, X. Toward tunable bandgap and tunable Dirac point in bilayer graphene with molecular doping. *Nano Lett.* 11, 4759–4763 (2011).
- Elias, D. C. *et al.* Control of graphene's properties by reversible hydrogenation: Evidence for graphane. *Science* **323**, 610–613 (2009).
- 20. Britnell, L. *et al.* Field-effect tunneling transistor based on vertical graphene heterostructure. *Science* **335**, 947–950 (2012).

ARTICLES

NATURE MATERIALS DOI: 10.1038/NMAT3518

- Britnell, L. *et al.* Electron tunneling through ultrathin boron nitride crystalline barriers. *Nano Lett.* 12, 1707–1710 (2012).
- 22. Yang, H. *et al.* Graphene barristor, a triode device with a gate-controlled Schottky barrier. *Science* **336**, 1140–1143 (2012).
- Li, X. S. *et al.* Transfer of large-area graphene films for highperformance transparent conductive electrodes. *Nano Lett.* 9, 4359–4363 (2009).
- Bhaviripudi, S., Jia, X. T., Dresselhaus, M. S. & Kong, J. Role of kinetic factors in chemical vapor deposition synthesis of uniform large area graphene using copper catalyst. *Nano Lett.* **10**, 4128–4133 (2010).
- Radisavljevic, B., Radenovic, A., Brivio1, J, Giacometti, V. & Kis, A Single-layer MoS₂ transistors. *Nature Nanotech.* 6, 147–150 (2011).
- Radisavljevic, B., Whitwick, M. B. & Kis, A. Integrated circuits and logic operations based on single-layer MoS₂. ACS Nano 12, 9934–9938 (2011).
- Loeser, A. G. *et al.* Excitation gap in the normal state of undoped Bi₂Sr₂CaCu₂O_{8+α}. *Science* 273, 325–329 (1996).
- Funahashi, R. & Shikano, M. Bi₂Sr₂Co₂O_y whiskers with high thermoelectric figure of merit. *Appl. Phys. Lett.* 81, 1459–1461 (2002).

Acknowledgements

We acknowledge the Nanoelectronics Research Facility and the Electron Imaging Center for Nanomachines (EICN) at UCLA for technical support of device fabrication and TEM characterization. We are grateful to I. Terasaki of Nagoya University for providing the BSCO samples. W.J.Y. acknowledges partial support by the National Research Foundation of Korea Grant funded by the Korean Government (NRF-2011-351-C00034). Z.L. is a visiting student from the Department of Physics, Peking University, sponsored by the UCLA cross-disciplinary scholars in science and technology (CSST) programme. H.Z. is grateful to the Camille and Henry Dreyfus Foundation for financial support through the Camille and Henry Dreyfus Postdoctoral Program in Environmental Chemistry (X.D.). X.D. acknowledges partial support by NSF CAREER award 0956171 (device fabrication and characterization) and ONR Young Investigator Award N00014-12-10745 (simulation). Y.H. acknowledges the NIH Director's New Innovator Award Program 1DP2OD007279 (TEM characterization and preparation of BSCO sample).

Author contributions

X.D conceived the research. X.D. and W.J.Y. designed the experiment. W.J.Y. performed most of the experiments including device fabrication, characterization and data analysis. Z.L. performed the simulations. H.Z. synthesized the graphene samples. Y.C. performed the TEM studies. Y.W. contributed to the preparation of BSCO flakes. Y.H. and X.D. supervised the research. X.D. and W.J.Y. co-wrote the paper. All authors discussed the results and commented on the manuscript.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to X.D.

Competing financial interests

The authors declare no competing financial interests.